Initial Retention by Vegetation of ^{131}I in Wet Deposition of Fallout

Contents

A8.1. INTRODUCTION

The purpose of this effort is to review the available literature on the initial retention by pasture vegetation of ^{131}I in wet deposition of fallout from the Nevada Test Site (NTS). Debris transported over distances in excess of 200 miles and deposited in rain during the months when cows consume pasture vegetation may have been responsible for the greater portion of the human intake of ^{131}I in the United States. The retrospective assessment of thyroid doses to the population requires knowledge of the interception and retention of fallout ^{131}I in wet deposition.

Some of the rainfall events were no doubt associated with thunderstorms and correspondingly high rainfall rates. At other times, wet deposition at low and moderate rainfall rates occurred. It would be useful to the dose assessment process to know the dependence of the retention factor on the rainfall rate and vegetation characteristics. Fallout near the NTS was generally due to dry deposition of relatively large particles and is not of interest in this review.

In **Section A8.2,** the context in which the initial retention factor is used is considered and its importance is illustrated. Models of transport of fallout to vegetation and of subsequent retention are presented to provide a basis for analysis of the available data. In **Section A8.3,** collected data on the nature of fallout ^{131}I in rainwater and of airborne fallout ^{131}I are reviewed. In **Section A8.4**, measurements of the retention of ^{131}I in wet deposition are summarized and evaluated. A tentative model relating the initial retention to vegetation density and total storm rainfall is presented. Data on retention of other fallout radionuclides in rain are reviewed, as are experiments with artificial sprays of radionuclides. Some available data on the retention of ^{7}Be are also reviewed. Conclusions drawn as a result of the review are presented in **Sections A8.5** and **A8.6.**

A8.2. IMPORTANCE OF INITIAL RETENTION FACTOR TO THE DOSE ASSESSMENT PROCESS

The transport of fallout ^{131}I from explosions at the NTS to members of the population can be modeled in a sequence of steps. For the principal exposure pathway these are:

- atmospheric transport and dispersion as the cloud segments move across the U.S.,
- wet and dry deposition of ^{131}I onto vegetation in the path of the cloud segments,
- consumption of contaminated pasture vegetation by cows,
- transfer of ^{131}I from the cows' feed to milk, and
- consumption by humans of cows' milk containing ^{131}I.

The first three steps define a similar sequence for the transport of ^{131}I to humans consuming fresh vegetables. In both pathways, the fraction of the ^{131}I in rain that is retained by vegetation enters directly into the calculation of the amount of ^{131}I consumed by humans.

The initial retention factor, a_v, is defined as the ratio of the ^{131}I concentration, pCi m^{-2}, on vegetation following a wet deposition event to the total ^{131}I deposited (also in pCi m^{-2}) during the event. This dimensionless parameter may be a function of rainfall rate, vegetation type, vegetation density, and the nature of the ^{131}I. The latter aspect, which includes both physical and chemical properties, is discussed in **Section A8.3.**

The transport of airborne ^{131}I to vegetation can be described mathematically using the following differential equation:

$$\frac{dC_v}{dt} = V_d \chi + a_v D_w - (\lambda_w + \lambda) C_v \qquad (A8.1)$$

where:

C_v	is the concentration, pCi m^{-2}, in vegetation
V_d	is the deposition velocity, m s^{-1}, to vegetation
χ	is the air concentration, pCi m^{-3}, normally at 1 m above ground level
a_v	is the dimensionless initial retention factor for wet deposition on vegetation
D_w	is the total wet deposition rate, pCi m^{-2} s^{-1},
λ_w	is the rate constant, s^{-1}, that describes removal by weathering processes
λ	is the radiological decay rate constant, s^{-1}

A similar equation can be written for the concentration of ^{131}I per unit mass of vegetation by dividing both terms of *equation A8.1* by the vegetation density or yield (Y, kg m^{-2}, dry weight).

The dry deposition velocity for elemental iodine (I_2) was found to be proportional to the vegetation density and a normalized deposition velocity, $V_D = V_d/Y$ was defined which reflected this dependence (Zimbrick and Voillequé 1969). In these measurements, the dry deposition onto the vegetation covering a specific area, not the total dry deposition (to vegetation and ground), was measured. This convention is also used in the definition of V_d in *equation A8.1.* The normalized deposition velocity has been recommended as a more useful parameter in dose assessment calculations (Hoffman 1977).

When the deposition velocity reflects the total rate of transfer (to both vegetation and ground) then a retention factor for dry deposition is also required. Chamberlain and Chadwick (1966) measured separately the activity of ^{131}I on both herbage and the underlying mat and soil due to deposition of I_2 released in field experiments. Chamberlain (1970) proposed a relationship of the type:

$$k = 1 - e^{-\mu Y} \qquad (A8.2)$$

where:

k	is the fraction of the radioactivity intercepted by the vegetation
μ	is a proportionality constant, $m^2\ kg^{-1}$, based upon experimental measurements
Y	is the yield or vegetation density, kg m^{-2} dry weight, as defined above.

For small values of Y ($Y < 0.3$ kg m^{-2}), the interception fraction is approximately directly proportional to the vegetation density. Chamberlain found a value of of 2.78 ± 0.14 $m^2\ kg^{-1}$ provided a good fit to the measurements of elemental iodine deposition. The measurements at Harwell include values of Y of ~ 0.7 kg m^{-2}; vegetation densities in the Idaho measurements were lower, ~ 0.2 kg m^{-2}, so the linear approximation was adequate.

Chamberlain also found that a variety of other experimental measurements gave similar values of μ when analyzed using the filtration model defined by *equation A8.2.* Releases of ^{89}Sr in a fine spray over grassland were performed by Milbourn and Taylor (1965). Analysis of the measured values of k yielded $\mu = 3.33 \pm 0.56$ $m^2\ kg^{-1}$. Because the ^{89}Sr in the solution was carrier free, the particles remaining after evaporation of the liquid were probably quite small. For similar releases of sprays and labeled particles 1 m in diameter (Chamberlain 1970), the best fit was provided by $\mu = 2.30 \pm 0.08$ $m^2\ kg^{-1}$. For labeled 30-μm diameter spores (Chamberlain 1967), a best-fit value of 3.08 ± 0.15 $m^2\ kg^{-1}$ was obtained. The similarity of values of μ for these physically different tracers (and for both dry deposition and spray application) suggested that reasonable estimates of retention could be made even if the radionuclide form was uncertain. For example, using the mean of the four estimates of μ with Y = 0.25 kg m^{-2} yields k = 0.51, while the extreme values of μ would predict 0.44 and 0.57 for the same vegetation density.

Miller (1979) confirmed that *equation A8.2* was representative of the initial retention of particles of various diameters used to simulate dry fallout. Those dry particles were spread over field vegetation plots and individual plants in small laboratory exposure chambers (Peters and Witherspoon 1972; Witherspoon and Taylor 1970, 1971). Particle sizes ranged from 1 to 44 μm (Witherspoon and Taylor 1970), 44 to 88 μm and 88 to 175 μm (Witherspoon and Taylor 1971), and 44 to 88 μm (Peters and Witherspoon, 1972). Several grasses as well as sorghum, squash, soybeans, and peanut plants were used in the studies. In a related paper, Miller (1980) analyzed the distribution of values of the retention factor and of the ratio of the retention factor to the vegetation density. The ratio (k/Y) was recommended for use for forage grasses in dose assessment calculations because of the lower variability. A median of 1.8 $m^2\ kg^{-1}$ and a geometric standard deviation of 1.6 were found. Seven of the ten experiments evaluated were for dry deposition of particles. The other three were the I_2 and spray results, cited above, that were used by Chamberlain.

Chamberlain's approach has been applied in the PATHWAY model being used to assess radiation doses from fallout near the NTS. Evaluation of field measurements of dry deposition of fallout particles between 80 and 260 miles downwind of the NTS led to the selection of $\mu = 0.39$ $m^2\ kg^{-1}$ for fallout dose assessments (Kirchner and Whicker 1983).

Lassey (1982, 1983, 1984) has also used Chamberlain's filtration model to describe the initial retention of radioactivity by vegetation. He has proposed an alternative to the commonly used exponential loss term for weathering processes (λ_w in *equation A8.1*) that is based on an extension of the filtration model.

It is reasonable to expect that the initial retention factor for wet deposition of ^{131}I will also be dependent upon the vegetation yield. Qualitative consideration of the wet deposition process suggests that other factors may also be important. The most prominent of these are the rainfall rate, the duration of the rainfall, and the rainfall sequence. High rainfall rates may result in lower net deposition on vegetation. The rain that falls at the end of a storm is less contaminated and may remove some ^{131}I that was deposited at the outset. Similarly, uncontaminated rain that falls after a wet deposition event may partially cleanse the vegetation. Thus, the fractional retention (a_v) in the wet deposition term of *equation A8.1* may actually be a complex function of many variables.

Different approaches have been used to address the effect of rainfall. In the model described by *equation A8.1* rainfall is just one of several possible mechanisms that contribute to the removal rate constant, λ_w. Peirson and Keane (1962) treated washoff by rain as the primary removal mechanism and assumed that the initial retention fraction was lowered when the rainfall rate increased. Their results are discussed in **Section A8.4.**

Horton (1919) considered the initial retention of rainwater by vegetation and developed the following expression:

$$a_v = \left(\frac{A}{P_s} + B \right) h \qquad (A8.3)$$

where:

a_v	is the fraction of the rainfall retained by the vegetation
A	is a constant equal to the rainfall storage capacity per unit height of vegetation (cm of rain per cm of vegetation)
P_s	is the amount of rain which falls during the storm (cm)
B	is the fraction of the rain that evaporates from the vegetation during the storm per unit of height of vegetation (cm^{-1})
h	is the height of vegetation (cm).

Horton estimated A and B using data collected at Seneca, New York, and some approximations about the rainfall pattern. For pasture grass and alfalfa, he estimated values of A of 4.2 x 10^{-4} and 8.3 x 10^{-4} (cm of rain per cm of vegetation) and values of B of 2.6 x 10^{-3} and 3.3 x 10^{-3} cm^{-1}, respectively.

The wet deposition rate, D_w, is itself complex because it reflects both in-cloud scavenging and washout of ^{131}I in the air nearer to ground level. Several symposia have been devoted to discussion of the relevant processes and field measurements to determine transport parameters (Engelmann and Slinn 1970; Semonin and Beadle 1977; Pruppacher et al. 1982; Georgii and Pankreth 1982).

For gaseous iodine species, the activity distribution between the ^{131}I in a raindrop and in the air around it is dynamic. Radioiodine entering the drop at one elevation may be lost from the drop at a lower elevation where the air concentration is lower. In some cases, wet deposition of ^{131}I will occur when there is no ^{131}I in the air at ground level. However, analysts of environmental measurements of wet deposition have been forced to correlate the wet deposition concentrations with ground level air concentrations because the concentrations at cloud level were not measured. The dimensionless washout ratio, W, is the ratio of the radionuclide concentration in rain, pCi kg^{-1}, to that in air pCi kg^{-1}, near ground level. This parameter can be used to define a wet deposition velocity, analogous to V_d. the washout ratios and wet deposition velocities that have been determined for fallout represent integrations of processes over times ranging from the duration of a single storm (perhaps 2 hours) to 1 week, a typical air sampler operating period. The field measurements frequently do not permit study of processes in the detail required to identify explicit dependencies upon rainfall rate or storm duration. Ground level measurements of washout ratios or wet deposition rates yield only relatively gross parameters that do not address the intricate details of the operative physical processes.

This is not to say that simple models are not useful. The natural integration of many variables that is inherent in a deposition velocity or washout ratio may be quite beneficial. Complex dependencies are smoothed and may counterbalance one another. For example, estimates of the wet deposition, D_w, using washout ratios include a dependence on the rainfall rate, p. If the retention factor, a_v, is inversely proportional to rainfall rate, it may be that the overall wet deposition term, $a_v D_w$, is approximately independent of p.

Direct correlation of deposition on vegetation with other field measurements of fallout may be an alternative to complex predictive modeling. Measurements of deposition of fallout were made using gummed-film collectors during the 1950s (Beck 1984). The samples were collected during a 24-h period and reflect both wet and dry deposition, removal, and decay. An equation similar to *equation A8.1* can be written for the activity on gummed film and evaluated on a daily basis. To use the gummed film results to estimate deposition on vegetation, it is necessary to know (a) the ratio of the dry deposition velocity for gummed-film to that for vegetation and (b) the ratio of the retention factors for wet deposition onto gummed film and vegetation.

Preliminary measurements (Beck 1986) suggest that the retention factor for both ^{7}Be, a dissolved species (Olsen et al. 1985) and ^{131}I in fallout from Chernobyl, partly particulate, decreases with increasing amounts of rainfall. Horton's conceptual approach, described above, appears quite appropriate for analysis of the retention of wet deposition by gummed-film. Approximate values for the storage capacity and evaporation fraction can be determined experimentally.

A8.3. PHYSICAL AND CHEMICAL FORM OF ^{131}I IN FALLOUT

Early measurements of radionuclides in fallout were frequently accomplished by gross beta counting the samples. Although gamma spectrometry measurements were performed earlier, routine determination of ^{131}I in samples did not begin until after the Windscale accident (Chamberlain and Dunster 1958). Early sample preparation procedures were not designed with ^{131}I in mind and in many cases, led to the loss of ^{131}I before the sample was counted. The gradual change in focus of studies of fallout in the United States is charted in a recently published review (Black and Potter 1986). Most information on ^{131}I in fallout has been obtained since above-ground testing at the NTS was completed. However, this fact should not greatly diminish the usefulness of those results for analysis of the behavior of ^{131}I in fallout generated at the NTS.

Three types of measurements of fallout characteristics are of particular interest. They are: (1) measurements of ^{131}I in wet deposition, (2) measurements of the solubility of ^{131}I in fallout particles, and (3) measurements of the partitioning of airborne fallout ^{131}I between particles and gaseous species. The first measurements are most closely related to retention of ^{131}I in wet deposition, but the other data provide supporting information

for inferences that must be made. Fortunately there are some data in all three categories and these are discussed below.

A8.3.1. ^{131}I in Wet Deposition

Four samples of rain and one of snow containing fallout ^{131}I were collected near Pittsburgh during November and December of 1962 and analyzed to determine the chemical and physical form of the radioiodine (Keisch and Koch 1963). Just prior to the start of these measurements, there was a large (>1000 kt) air drop test at Johnston Island. During the period there were five atmospheric tests (20 to 1000 kt) in the Soviet Union, four underground tests at the NTS, and two missile tests (<20 kt and <1000 kt) at altitudes of tens of kilometers in the Pacific (Reiter 1978). An average of 51 ± 17% of the ^{131}I activity was in the liquid phase, which was operationally defined by passage through a filter with a pore size of 1.2 μm. (Unless otherwise indicated, the mean, M, and the sample standard deviation, s, of measurement results are given in the form M ± s.) The range of the liquid phase fraction for the five samples was 23% to 65%. The solids were described as "fine suspended particles" or "settled particles"; most of the particulate activity (an average of 76 ± 14%) was in the latter category. All of these particles must have exceeded 1.2 μm in diameter.

After the particles were separated from the rainwater, they were exposed to deionized water and gently agitated to determine the further availability of the ^{131}I in the fallout particles. An average of about 8.6 ± 5.6% of the ^{131}I activity was leached from the particles by deionized water in 1 hour; values for six samples (four of fines and two of settled particles) ranged from 3 to 15%.

Measurements of the chemical state of the ^{131}I in the liquid phase of five precipitation samples were also made. An average of 52 ± 15% was determined to be present as iodide or iodine, 37 ± 15% was identified as iodate, and 11 ± 9% was found to be periodate. Similar measurements were made to determine the chemical state of $^{131}1$ that was subsequently leached from four samples of the particulate fraction. The distribution of chemical forms of ^{131}I leached from particles deposited in rainwater was similar to that found in the liquid phase of the rainwater. The results for the iodide/iodine, iodate, and periodate fractions were 65 ± 21%, 23 ± 16%, and 10 ± 12%, respectively.

The development of analytical methods for these measurements is described in an earlier report by the same authors. Their original measurements were made on particles collected within about 2 miles from ground zero following the Sedan Test (Koch and Keisch 1962). They found that results of leaching with deionized water and an acid solution determined to resemble gastric juice were similar. Additional measurements of the long-lived isotope ^{129}I in fallout were planned by the same investigators, but a report of that work has not been found.

Indirect evidence of the nature of ^{131}I in wet deposition is provided by the measurements of washout ratios. Measurements of tropospheric fallout from Russian nuclear tests in 1961 yielded washout ratios of 420 for ^{131}I, 480 for ^{140}Ba-La, and 500 for ^{95}Zr-Nb (Peirson and Keane 1962). These results were comparable to those from measurements of long-lived fallout originating in the stratosphere during the previous year. Washout factors of 560 and 520 for ^{137}Cs and ^{144}Ce-Pr, respectively, were reported. The similarity of the ^{131}I washout ratio and those for the particulate radionuclides suggests a common origin, namely fallout particles. Washout factors for a much wider range of conditions are given in the review by Engelmann (1970). Only one value is given for ^{131}I; washout ratios of 100 to 2700 were measured for snow containing ^{131}I from the Cabriolet venting at the NTS. Engelmann (1970) also quotes work by Bradley who analyzed beta activity washout ratios in Illinois from 1962 to 1965 and found a nominal value of 490 with a slight dependence upon total monthly precipitation (P, cm): $W_{beta} = 490\ p^{-0.026}$.

The atmospheric cleansing effect is shown more dramatically in measurements of ^{90}Sr washout (Krey and Toonkel 1977). The ^{90}Sr washout ratio at Seattle was between 1964 and 1967 and was proportional to $P^{-0.3}$. The combined data for three cities (Seattle, Fayetteville, and New York) showed a proportionality to $p^{-0.17}$. For the three sites, the value of W for ^{90}Sr for P = 1 cm was 969, about double that found for gross beta activity in Illinois. During the 1964-1967 period, most of the ^{90}Sr would have come from the large stratospheric inventory built up prior to January 1963. Above ground nuclear testing by the United States and the Soviet Union was stopped by treaty in 1963, but tropospheric explosions occurred in the northern hemisphere throughout 1962. There were, however, six Chinese nuclear explosions during the period when the measurements were made (Reiter 1978).

A8.3.2. Leachability of ^{131}I in Fallout Particles

Airborne particulate material in the Pittsburgh area was collected on glass fiber air filters (Gelman) and used to measure the leachability of ^{131}I into deionized water (Keisch and Koch 1963). The filters were highly efficient for particles as small as 0.05 μm. Weekly samples were collected between November 21, 1962 and January 2, l963. After leaching, the liquid phase was operationally defined by filtration using a filter with a 1.2-μm pore size. An average of 29±5% of the total ^{131}I activity was found in the liquid phase after 4 hours of gentle agitation in deionized water. Most of the ^{131}I activity entered the liquid soon after contact. After the first hour of leaching, an average of 23±4% of the ^{131}I activity was found in the liquid phase.

The chemical form of the leached activity in five samples was identified as 61±18% iodide/iodine and 32± 17% iodate. The average periodate fraction can be estimated by difference to be about 7%, but three of the five analytical results for periodate were below the detection limits. There is a definite similarity between these species distributions to those in the liquid phase of rainwater and to those found after leaching particles brought down by rainwater (**Section A8.3.1**). The results suggest that ^{131}I in the liquid phase of the precipitation samples may have been

due to leaching of ^{131}I from particles during droplet formation and while the precipitation fell.

Other measurements of the solubility of ^{131}I in fallout particles were less oriented toward leaching in raindrops. In two samples, Perkins (1963) found that 42% and 44% of the ^{131}I was leached from particles by a basic solution (pH 12) in a blender in 2 minutes. The filter had a pore size of 2 μm so some of the liquid fraction may have been small particles. Destruction of large particles no doubt occurred in the blender, which further complicates interpretation of this result. The two iodide/iodine fractions were about 57 and 66% and iodate accounted for about 38 and 29% of the leached ^{131}I in these samples, respectively. The periodate fraction was less than 5% in both samples (Perkins, 1963).

A8.3.3. Forms of Fallout ^{131}I in Air

In a series of measurements made during the intensive periods of bomb testing in 1961 and 1962, Perkins and his associates made regular measurements of airborne ^{131}I. The fraction of the ^{131}I that was in particulate form ranged from about 1%, in a single case, to more than 90% and nearly always exceeded 10% (Perkins et al. 1965). The mean particulate fraction can only be estimated from points on a greatly reduced figure that shows the measured particulate fractions (Perkins et al. 1965); it appears to be ~ 0.5. The fraction of the gaseous fallout ^{131}I present as I_2 or HI was estimated to be less than 10%; however, the number of fallout measurements when the gaseous species were separated was not given. The remainder of the gaseous fraction was presumed to be in organic form (Perkins 1963, Perkins et al. 1965).

In a series of measurements at Brookhaven National Laboratory (BNL) during the last 5 months of 1962, Hull (1963) found an average of 65% of the ^{131}I was associated with particles. The particulate fraction ranged from 21% to 82% during the measurement period (Hull 1963). Eggleton et al. (1963) reported an average particulate fraction of 75% during the fall of 1961. The distribution of the gaseous species cannot be determined from the abstract and the entire paper was not published (Eggleton et al. 1963).

Measurements of fallout ^{131}I from Chinese weapons testing in 1976 showed distributions of airborne activity that were similar to the earlier measurements. The samples were collected in the vicinity of a nuclear power plant in New Jersey. Following the test, fallout ^{131}I concentrations in air greatly exceeded those due to facility releases of ^{131}I (Voillequé 1979). Fallout ^{131}I was measured at five sites during a 2-month period; eight weekly samples were collected at each location. About 50 to 60% of the total airborne ^{131}I activity was present as particles during the first 5 weeks following arrival of the fallout. During the last 3 weeks, the particulate fraction decreased to about 30% of the total. The fraction of the gaseous ^{131}I in organic form averaged 35% during the first week and about 40% during the second and third weeks. The gaseous iodine was predominantly in organic form during subsequent weeks with mean values for all sites ranging from 44% to 100%. The amount of ^{131}I in the I_2 or HOI) fraction was often below the detection limit so the total gaseous ^{131}I activity and the fraction that was in organic form were both indeterminate. However, the observed trends indicate that the organic fraction gradually increased with time after detonation.

Measurements made in Germany (Riedel et al. 1977) following the same test showed an initial particulate fraction of 0.72. The fraction associated with particles declined, although not monotonically, to 0.54 after 5 weeks. The gaseous fraction was collected using charcoal; no attempt was made to determine the distribution of the gaseous iodine forms.

Fallout ^{131}I from a Chinese test in September 1977 was observed in the midwestern United States within 5 days of the detonation. Two measurements of the particulate fraction at each of two locations yielded values between 0.54 and 0.59 during the first 3 weeks after the explosion. Subsequent airborne ^{131}I concentrations were too small to permit evaluation of changes in chemical form of the fallout and measurements were discontinued (Keller et al. 1982).

Examination of the plot of the particulate fraction of airborne ^{131}I in 1961 and 1962 (Perkins et al. 1965) shows several distinct declines in that quantity following peaks that presumably indicate arrival in Richland of fresh fallout from a recent test. However, testing was so frequent in those years that mixtures of fallout ^{131}I from a variety of tests would tend to obscure any trends related to the age of the fallout.

The measurements of ^{131}I in particles in precipitation (Keisch and Koch 1963) showed that half of the ^{131}I activity in wet deposition was associated with particles larger than 1.2μm in diameter. Although diameters of the settled particles were not measured, the description suggests that these particles were visible to the unaided eye. Peirson and Keane (1962) reported upper limit diameters in the range of 1 to 4 μm based upon microscopy and autoradiography. Approximately 50 to 80% of fallout particle beta activity was found to be associated with particles with diameters greater than 1 μm (Lockhart et al. 1965). The observed gradual reduction in the particulate fraction probably reflects removal of larger particles by gravitational settling (G1asstone 1964) and perhaps by precipitation scavenging (Gatz 1977; S1inn 1977). Fallout particles with diameters greater than 25 m have predicted residence times of less than 1 day, even when injected at an elevation of 50,000 feet (G1asstone 1964).

Studies of beta activity of fallout particles from a high altitude burst, which had diameters between 2 and 20 μm, showed that the activity was approximately distributed uniformly throughout the particle volume (Benson et al. 1965a). No measurements of the distribution of ^{131}I were reported. Individual particles exhibited widely varying activity concentrations and such differences might be even greater for near surface explosions. The same authors performed radiochemical studies (Benson et al. 1965b) but did not detect ^{131}I (or ^{103}Ru or ^{137}Cs) in particles that they studied. The ^{95}Zr-Nb and ^{140}Ba-La peaks interfered with analyses of these nuclides (a NaI(Tl) detector was

used). The activity distributions and fractionation data were not available for those nuclides. Other studies of fallout particles (Krey and Fried 1965; Crocker et al. 1965; Friedlander and Pasceri 1965) provide precious little information about ^{131}I and its incorporation into fallout particles. One may speculate, using the general scheme provided in Friedlander and Pasceri (1965) and a few bits of information on ^{132}Te and ^{131}I, that more than half the ^{131}I would be associated with particle diameters less than about 20 μm and be found principally in fallout that becomes widely distributed. As a volatile element, iodine might be expected to condense on surfaces rather than be distributed throughout the particle volume, but this has not been demonstrated.

A8.4. MEASUREMENTS OF RETENTION BY VEGETATION OF WET DEPOSITS OF FALLOUT ^{131}I AND OTHER RADIONUCLIDES

Field measurements of total wet deposition and of the activity present on vegetation after rainfall have permitted estimates of the wet deposition retention parameter a_v for ^{131}I and other fallout radionuclides. Artificial applications of radionuclides to vegetated areas have also yielded estimates of a_v. Measurement programs that specifically studied ^{131}I are of course of greatest interest, but results for other radionuclides that define the initial retention of fresh fallout particles are also of great interest. Results from experiments when radionuclides are dispersed at ground level are inherently less valuable for assessing the retention of particulate ^{131}I, although, as indicated in **Section A8.2**, Chamberlain found the filtration model was consistent with results for various physical forms and modes of application.

A8.4.1. Retention by Vegetation of Fallout ^{131}I

Using daily values of the concentrations of ^{131}I in air, rain, and vegetation for the British Isles during the fall of 1961, Chamberlain and Chadwick (1966) evaluated the dry deposition velocity and the wet deposition retention factor. Because there were alternating periods when wet and dry processes were predominant, good estimates could be obtained for both parameters. The frequently observed 5-day effective half-life for ^{131}I on vegetation was used in the calculations. The means and standard errors obtained by least squares fitting procedure were V_d = 0.054 ± 0.009 m/s and a_v = 0.51 ± 0.10. The three largest daily rainfalls occurred in late October and were in the range of 0.8 to 1.4 cm.

At Chilton (UK) measurements were made of fallout ^{131}I from Russian tests conducted in 1961 (Peirson and Keane 1962). Weekly average data obtained during 2.5 months were used to estimate parameters for wet and dry deposition and removal by rainfall. The dry deposition velocity was estimated to be about half that found by Chamberlain and Chadwick. In the analysis, it was assumed that the retention factor was decreased by increased weekly rainfall (P_w) according to:

$$a_v = (1 - m P_w) \qquad (A8.4)$$

where

m was a constant to be determined.

Washoff of activity by subsequent rains, also assumed to be proportional to the weekly rainfall, was considered to be the principal removal mechanism. The value of m determined for ^{131}I was 0.015±0.013 (week per mm of rain). The best estimate of the ^{131}I washoff factor was even more uncertain, 0.020±0.028 per mm of rain. Using the value of m stated above, the values of a_v for 9 weekly periods were found to range from 0.48 to 0.99. Measured rainfalls varied from 0.1 to 2.9 cm and 1 week passed without any precipitation. The mean of the computed values of a_v was 0.78 ± 0.18.

One would not expect that these results, which substantially overlap in time, would be so different from the results presented by Chamberlain and Chadwick (1966). Part of the difference lies in the assumptions that were made by Peirson and Keane. The effect of dry deposition on the precipitation collector was not evaluated. Removal mechanisms other than washoff were not considered, so removal was somewhat underestimated. It may also be that the use of weekly average values, rather than day by day results, skewed the results. There are of course relatively large uncertainties in the estimates of V_d (0.26 ± 0.18), m, and the washoff factor. Those uncertainties may be partly due to the approach taken in data evaluation.

It is recognized that other processes may influence the results of such field experiments. Rainsplash of previous deposits onto vegetation and uptake of ^{131}I from the soil are possible confounding factors. In the fall of 1961, the estimated ^{131}I wet deposition rate was about 300 pCi m^{-2} d^{-1}. In about a month, an equilibrium deposit of about 3500 pCi m^{-2} would be achieved. If all the activity were in the top cm of soil, only about 1 pCi m^{-2} would be expected due to soil uptake by vegetation in equilibrium with the soil (NRC 1977). This is substantially lower than the deposition rate. A rainsplash transfer fraction of 1% of the soil activity would not greatly influence the estimates for vegetation. If significant rainsplash occurred, its effect would have been to raise the apparent initial retention factor.

Hull (1963) reported measurements of ^{131}I concentrations in air, precipitation collectors, vegetation, and milk at BNL during the period August to December 1962. Chamberlain and Chadwick's values of V_d = 0.05 m s^{-1} and a_v = 0.5 were used with the BNL measured weekly average concentrations of ^{131}I in air and deposition collectors to predict concentrations of

^{131}I on grass. The fit of predicted values to measured concentrations of ^{131}I on grass was apparently considered satisfactory, and no attempt was made to determine best-fit values of V_d and a_v using the BNL data. The rainfall times and rates were not given. The deposition totals may not have been corrected for the effect of dry deposition, so a best-fit value for av might be slightly greater than 0.5.

During this same period, measurements of ^{131}I in environmental samples at the Studsvik research center in Sweden were also underway. Data on ^{131}I in air, rain, and milk were reported by Bergström (1967) and Bergström and Gyllander (1969). Specific measurement results were not presented for vegetation and ^{131}I retention, but values of a_v of 0.3 for light rains and 0.1 to 0.2 for heavy rains were stated to be "in agreement with the measurements of fallout iodine in Sweden" (Bergström 1967). About half of the rainfall rates during the measurement period were in the range 0.5 to 2.0 cm d^{-1}; the remainder were lower.

Fallout from more recent atmospheric weapons testing has complicated attempts to monitor the behavior of ^{131}I released from nuclear power stations. The environmental concentrations of ^{131}I from fallout episodes are much greater than those due to station effluents. These occurrences have led to collection of data on fallout ^{131}I. In the midwestern United States during June and July 1973, several values of a_v were determined. Grass and precipitation samples were collected following rainfall events. Concentrations of ^{131}I in grass prior to the wet deposition were frequently at the detection limit, so corrections for removal of previous deposits were generally not required. When necessary, extrapolation of vegetation concentrations was used to estimate the concentration prior to the rain. An effective removal half-life of about 5 days was generally observed. Daily rainfall totals ranged from 1.0 to 2.9 cm and the estimated retention factors ranged from <0.09 to 0.52. Two of the lower values of a_v (both < 0.09) were associated with rainfalls of 2.2 and 2.9 cm. The highest value (0.52) was observed for a rainfall of 1.8 cm. Detailed data on rainfall rates during storms were not reported (Weiss et al. 1974).

Radioiodine measurements in the environs of a reactor the following year were again interrupted by Chinese fallout. Four measurements of the initial retention of ^{131}I in wet deposition ranged from 0.11 to 0.27. In this case, the two lowest values (0.11 and 0.18) were associated with the smallest rainfalls (trace to 0.5 cm), and the highest initial retention fractions (0.25 and 0.27) were for a total rainfall of 1.3 cm.

At the reactor site in New Jersey in 1976, no reliable data on wet deposition were available after arrival of the fallout. The published analysis (Riedel et al. 1977) of measurements made in Germany following the same test assumed $a_v = 0.2$ based on Regulatory Guide 1.109 (NRC 1977). Two effective retention half-lives were examined: 3.9 and 5 days. Dry deposition rates were derived using these assumptions and the assumption that the aerosol deposition velocity was one-tenth that for gaseous iodine. This approach to the data was clearly arbitrary. It may be that the authors felt the number of vegetation samples (one a week) was too small to permit a detailed evaluation of wet deposition. The approach may also have been influenced by the low rainfall. It averaged 0.86 cm/week and in 6 of 9 weeks the total rainfall was less than 1 cm.

Measurements made during the summer of 1977 at a site on the Mississippi River also showed the presence of Chinese fallout (Voillequé et al. 1981). Measurements permitted ten estimates of the initial retention parameter a_v. The range of the estimates was large, 0.1 to 0.9; the mean retention factor was 0.35+0.28. The total rainfalls that carried wet deposition to the ground ranged from 1.0 to 5.8 cm, but these accumulations occurred over varying periods. Descriptions of the rainfall patterns were somewhat more detailed than in other publications, but variations within individual storm periods which lasted from several hours to several days, were not reported.

The need for detailed rainfall data seems clear from the point of view of wet deposition process modeling. Rainfall rates during storms can vary by more than an order of magnitude and scavenging processes are expected to be affected by those changes (S1inn 1977). The retention factor may be lower for higher rainfall rates. More washoff may occur (although it must be said that the importance of washoff as a removal process has not been unequivocally demonstrated). On the other hand, a retrospective study of fallout deposition will be limited in sophistication by the data collected at that time. It is unlikely that precipitation rates during storms will be available; it is probable that storm or daily total rainfalls will be used. So it may be that average retention factors for whole storms, or for 24-h periods in which a specific quantity of rain fell, are the most relevant for the problem at hand.

It is encouraging that Huff (1965) found that the best correlations of beta activity deposition were with rainfall volume, not with duration or rate. Analysis of 15 storms also showed that a single station could be used to predict the deposition of gross beta radioactivity within 10 to 12 square miles within an average error of 22%, however, the average error was as large as 35% in one-third of the storms.

It has long been known that rainfall concentrations decrease during the course of a storm (Weiss 1953). Recent automated measurements of metals in sequential rainfall samples show that the concentrations in rain decrease substantially at first, but are relatively constant after 2 to 3 mm of rain have fallen (Kins 1982). Hence, the largest concentrations are deposited when retention may be most likely, before saturation of the plant surfaces. Horton's estimates indicate that the storage capacity of tall (30-cm high) grass would be reached in less than a minute during a well organized storm with an average rainfall rate of 1.5 cm/hr. However, Burgy and Pomeroy (1958) indicate that the storage capacity is satisfied in increments during a storm (Bu58). More information about binding mechanisms and rates is needed to evaluate this aspect.

In the following discussion, a synthesis of the existing data on initial retention of ^{131}I in fallout is attempted and a ten-

tative model is developed. *Figure A8.1* contains the results of measurements of the initial retention of ^{131}I in Chinese fallout by pasture vegetation in the midwestern United States. Some of the values for a_v for nearly the same densities vary by more than a factor of 4. Also shown for comparison is the predicted line from Chamberlain's best fit to the 30-μm *Lycopodium* spore data. Average values of a_v for three ranges, each of which includes five to seven results for similar vegetation densities, are shown as bars in the figure. These means are not in good agreement with that model and the sample standard deviations, indicated by the vertical bars, are quite large. The single measurement for $Y > 0.2$ kg m^{-2} is a factor of two below the curve for 30-μm spores. The results of the two analyses of tropospheric fallout in the UK are not shown because vegetation densities were not reported.

Figure A8.2 shows the initial retention factor as a function of total storm rainfall (P_s, cm). As in *Figure A8.1*, the circles are the data from the midwest. The hatched area at the left encompasses the range of storm rainfall totals estimated from Chamberlain and Chadwick (1966); the reported average retention factor of 0.5±0.1 was based on daily grass samples. During some weeks, rain fell for several consecutive days; the maximum value of P_s is estimated to be <2 cm. Grass cut after the first part of an extended rainfall would have experienced a lower value of P_s, but not lower than 0.2 cm. The uncertainty associated with the mean retention factor is not reflected by the hatched area. The uncertainties appropriate for the U.S. data are generally larger (~ 40%). Only the best estimates are shown in the figures to improve legibility. The estimates made by Peirson and Keane are even more uncertain (as they are based on m = 0.015±0.013). The number of storms at Chilton was estimated using data (Chamberlain and Chadwick 1966) and mean values of P_s for the Chilton data were estimated. The average retention factors in Peirson and Keane (1962) are plotted as open squares in *Figure A8.2* using those estimates.

Incorporating the estimates (in an admittedly approximate way) does clarify the overall pattern. While the range of values of a_v for $P_s < 2$ cm is large, part of the variation is undoubedtly due to differences in vegetation density. Two patterns can be seen. One is a gradual linear decrease of a_v with P_s, as assumed in Peirson and Keane (1962); the other is the sharp decrease for P_s <2 cm, followed by a much slower decrease in a_v for P_s >2 cm. This latter pattern is that predicted by Horton (1919) and a plot of that type of model with assumed values of A and B included in *Figure A8.2*. However, neither type of model satisfactorily predicts all of the experimental results.

For modeling purposes, a combination of the filtration model and Horton's approach could be used. First, we define a normalized retention factor (a^*_v, m^2/kg)

$$a^*_v = \frac{a_v}{Y} \qquad (A8.5)$$

This approach has been suggested by Miller (1980), although it is clear from *Figure A8.l* that this normalization will not greatly reduce the variability in the available data for ^{131}I. Based on data in Chamberlain and Chadwick (1966) and a range of wet to dry weights of 1/4 to 1/3, dry vegetation densities in the late fall in the UK seem likely to have been in the range 0.08 to 0.15 kg m^{-2}. The distribution of values of a_v is expected to be similar to that for a^*_v in *Figure A8.2.* This suggests that a^*_v will depend upon the total rainfall from a storm in a manner similar to that proposed in Horton's model. That dependence could take an alternative form:

$$a^*_v = \frac{S}{P_s} + E \qquad (A8.6)$$

where:

S is the rainfall storage capacity per unit areal density of vegetation

E is the in-storm evaporation fraction per unit areal density of vegetation

To determine the fractional initial retention by pasture vegetation of ^{131}I in fallout from a particular storm, substitute the storm rainfall total, P_s, and vegetation density, Y, into *equation A8.6*.

$$a_v = \left(\frac{S}{P_s} + E \right) Y \qquad (A8.7)$$

Values of S and E are estimated from the data and assumptions discussed above to be 1.6 and 1.3 m^2 kg^{-1}, respectively. Although E in *equation A8.6*, and B in Horton's original formulation shown in *equation A8.3*, are assumed to be constants, the evaporation fraction certainly depends upon the air temperature and relative humidity and thus on the time and duration of the storm.

A8.4.2. Retention by Vegetation of Other Radionuclides in Fallout

To the extent that ^{131}I in fallout is associated with particulate material, the behavior of other radionuclides incorporated in fallout particles should be similar. In fresh fallout, 60 to 90% of the ^{131}I has been found in the particulate fraction and the gaseous ^{131}I is mainly in inorganic form. If the particulate fraction were 75%, for example, about 20% of the total would be expected to be I_2, HI, or HOI, and only about 5% would be expected to be in organic form. The behavior of particulate radionuclides should be reasonably representative of ^{131}I transport soon after detonation.

As the time after detonation increases, the larger particles are removed from the atmosphere and less airborne ^{131}I is associated with particles. At the same time, the less reactive organic iodides become an increasingly larger fraction of the gaseous fraction. Two months after a detonation, the airborne iodine species distribution would be quite different from that cited above. Only about 30% of the ^{131}I would be associated with particles and the organic iodides would comprise about 50 to 60% of the total with the remaining 10 to 20% as inorganic gases. At

Figure A8.1. Initial retention of ^{131}I in Chinese fallout in the Midwest. Data from Voillaqué et al. (1981) and Weiss et al. (1974).

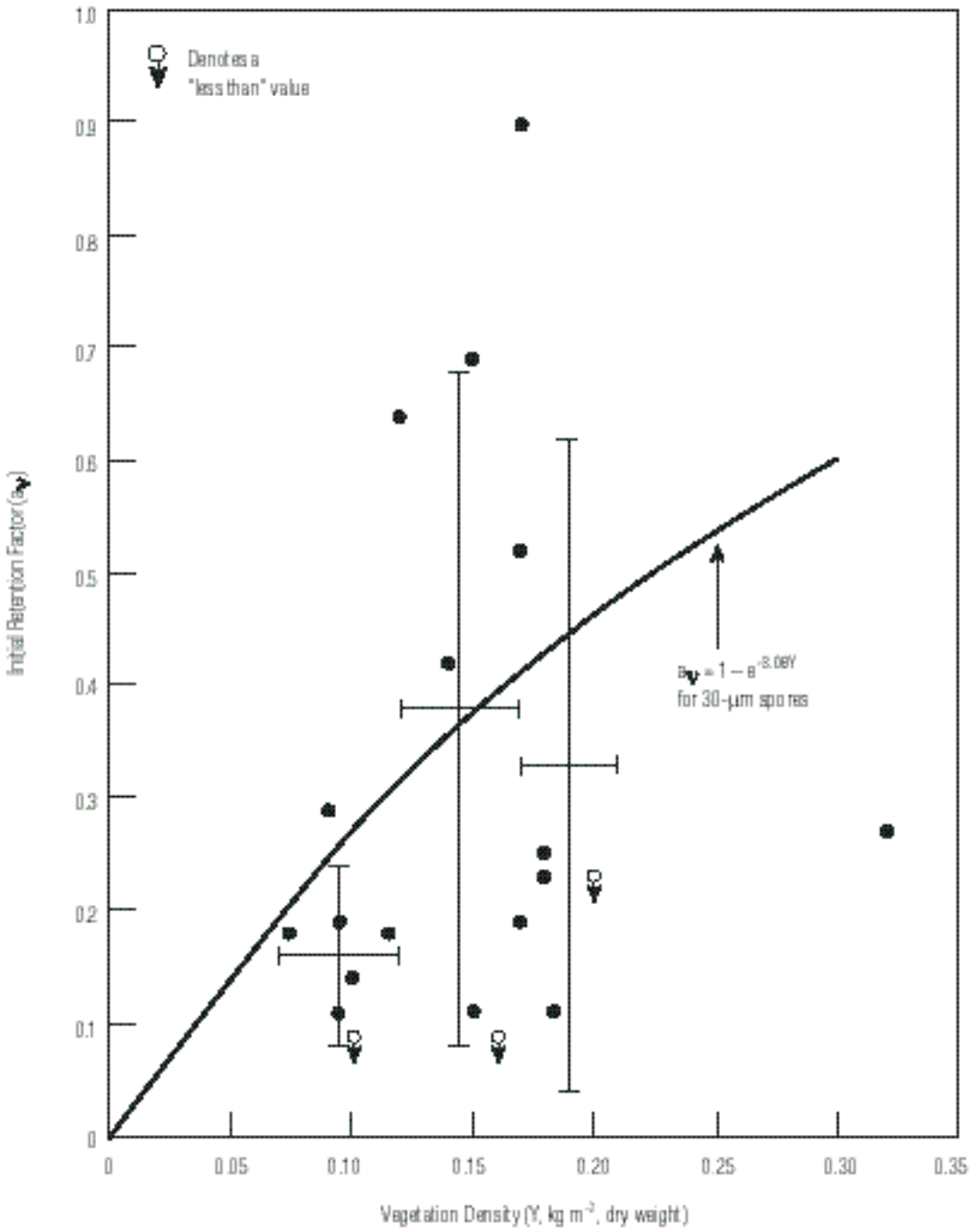
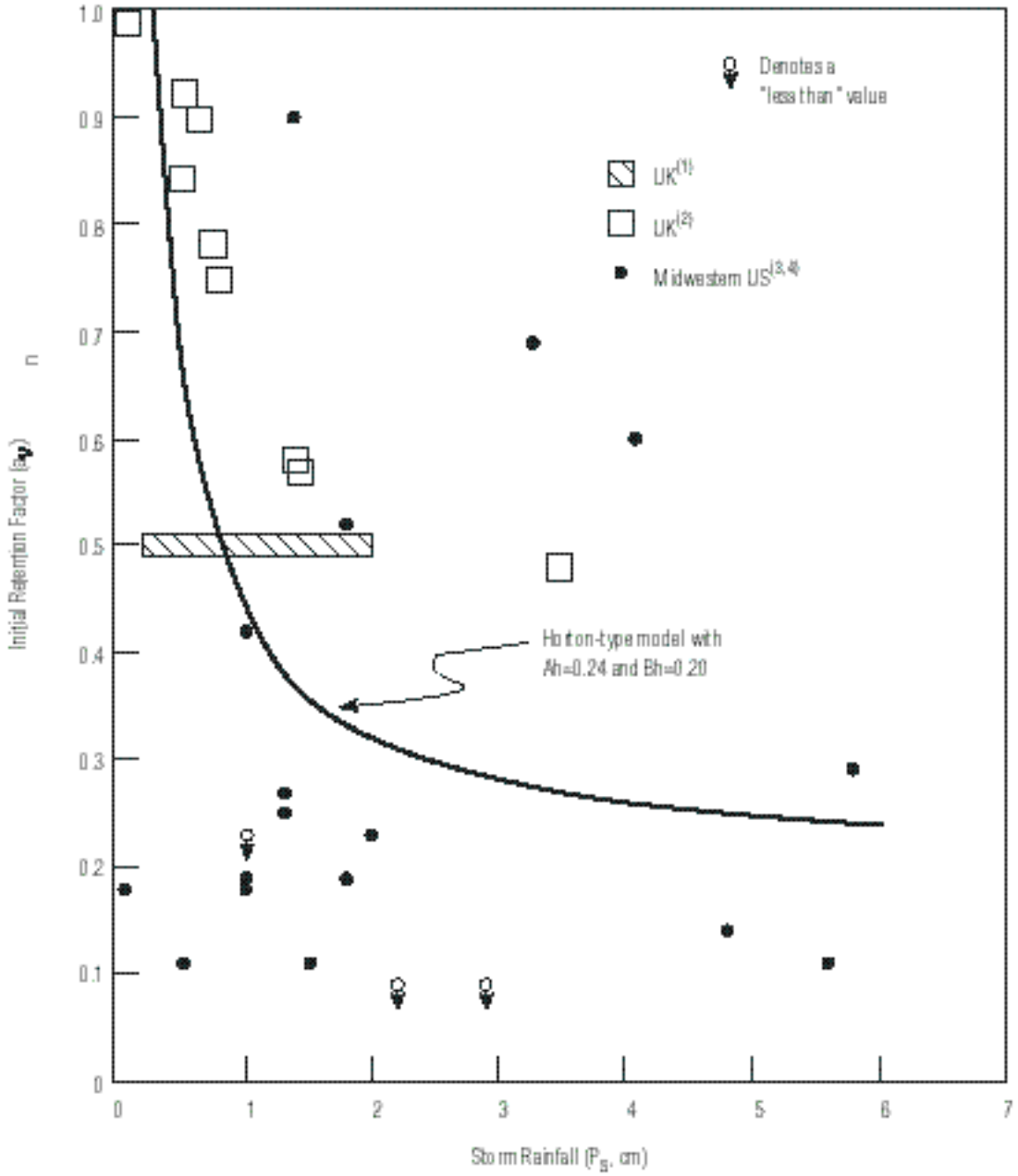


Figure A8.2. Dependence of initial retention of fallout ^{131}I on storm rainfall. Data from (1) Chamberlain and Chadwick (1966), (2) Peirson and Keane (1962), (3) Voillequé et al. (1981) and (4) Weiss et al. (1974).



that and later times, the behavior of residual particulate fallout radionuclides would be representative of less than 1/3 of the total ^{131}I. The principal concern for the NTS fallout is the distribution during the first 10 days following a detonation.

Another distinction is important: many of the measurements of fallout particles (mainly ^{90}Sr and ^{137}Cs) were made after the cessation of atmospheric nuclear testing in the northern hemisphere. The fallout at the later times was derived from the substantial stratospheric inventory established earlier. Only periodic testing by the Chinese has provided recent opportunities for measuring fallout from low level air bursts. It is also noteworthy that the stratospheric particles were found to be quite small, <0.3 μm in diameter (Drevinsky and Pecci 1965; Loysen 1965), although measurements of fallout in the troposphere indicated attachment of fallout particles to naturally occurring aerosol particles or agglomeration to form particles with diameters of about 0.3 to 2 μm (Friedlander and Pasceri 1965; Lockhart et al. 1965). However, even the composite particles are smaller than would be expected within a few days after an explostion at the NTS.

Too little is known of the interactions on the plant surface that result in attachment of radionuclides to predict what the effect of the particle size difference would be. However, caution is advised, both because of that difference and because of the presence of other airborne iodine species.

During the period of tropospheric fallout in the fall of 1961, Peirson and Keane found that the particulate radionuclide ^{140}Ba-La behaved in a manner quite similar to ^{131}I. Estimated mean dry deposition velocities and washout ratios were nearly identical for the two nuclides. The average initial retention fraction for ^{140}Ba-La was estimated to be 1.0±0.3 compared with 0.80±0.17 for ^{131}I using the approach described above.

As indicated above, most studies of ^{90}Sr and ^{137}Cs were of older fallout. Ward et al. (1965) measured the wet deposition of ^{137}Cs and activity on pasture vegetation and cut alfalfa. Deposition on alfalfa from six storms in May and June of 1964 was measured. Five of the evaluations yielded initial retention factors between 0.26 and 0.83, the mean was 0.60±0.21. The sixth result was about 1.8, indicating experimental difficulty or that the predicted removal of previous deposits was less than had actually occurred prior to the storm. During the spring and summer of both 1963 and 1964, the cumulative ^{137}Cs in cut alfalfa hay was measured, as was the wet deposition of ^{137}Cs during the 5- to 7-week growth period. The concentration of ^{137}Cs in air was not reported and neither dry deposition nor weathering was considered. Inclusion of weathering changes the published estimates of av for alfalfa by more than a factor of 2, and 2 of the revised values exceed 1. However, in a thesis (Wilson 1968) cited by Anspaugh (1987), it is stated that their original deposition measurements were found to contain only about half as much ^{137}Cs as was found by an alternative measurement technique, so this would lower computed values of a_v. During 4 of 6 growing periods, the weekly precipitation averaged less than 0.3 cm, so dry deposition was no doubt responsible for most of the activity found in the alfalfa.

Later measurements of retention of wet deposition by alfalfa indicated that washoff of ^{137}Cs occurred during 3 of 7 periods (Wilson et al. 1967). However, the sampling and analysis did not distinguish between washoff and other removal processes. There is also uncertainty about the amount of dry deposition. Dry deposition was shown to be an important process by comparing covered and uncovered vegetation areas, but its effect on the data for exposed alfalfa was not analyzed. Reference is also made to procedural difficulties in the wet deposition measurements. An improved procedure was developed that involved scrubbing of the collector surface to assure collection of all of the deposit in the cation-exchange bed into which precipitation was funneled. This change presumably corrected the problem cited above.

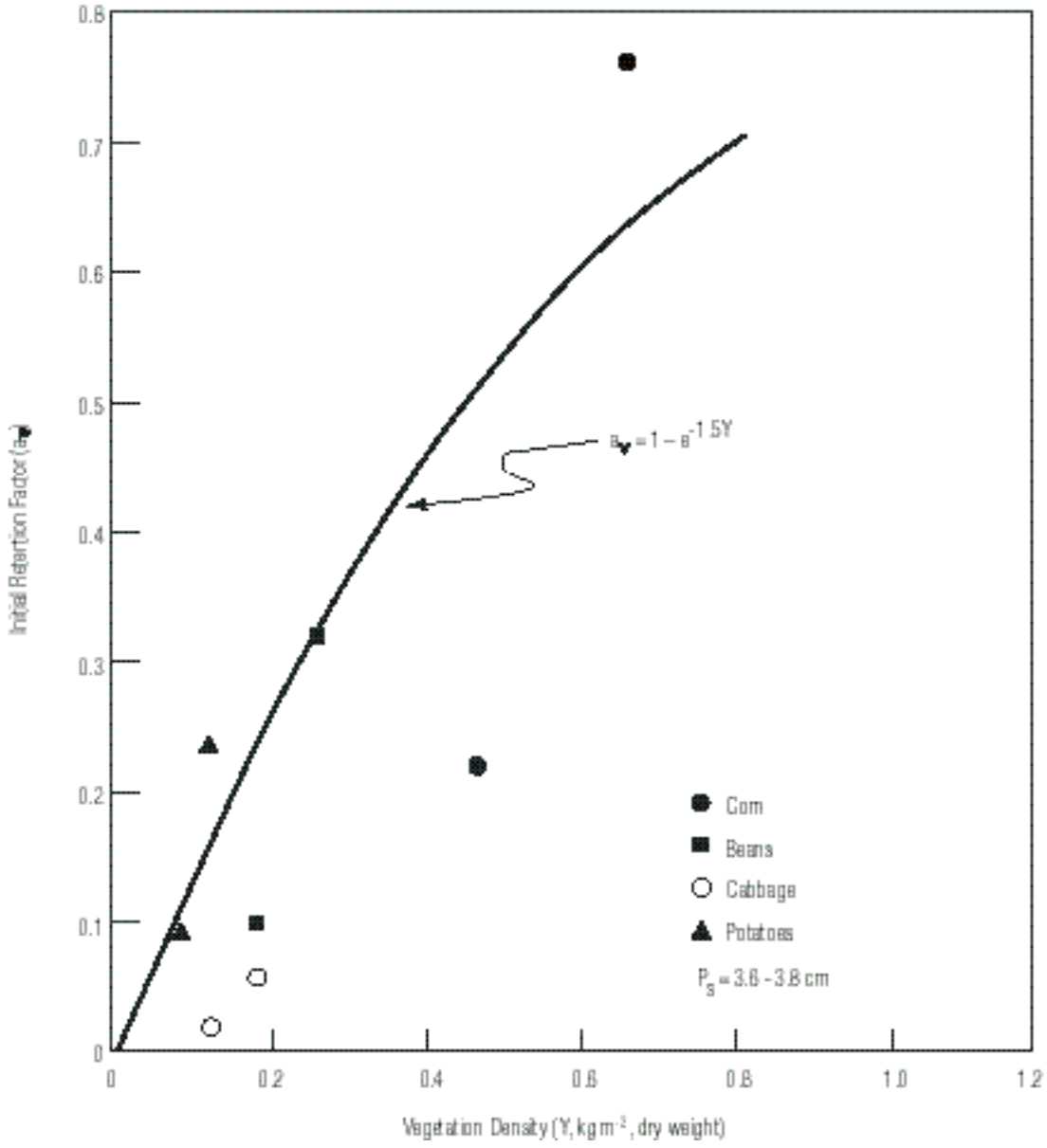
A detailed study of retention by wet deposition was conducted in Florida in the spring of 1962 (Menzel et al. 1963). Retention of ^{90}Sr on low density young plants was less than 10%. As the vegetation grew, the initial retention increased. Vegetation samples were collected before and after each rain, but yields were not stated at the times of the rainfalls. The initial retentions for four crops during two rainfalls of nearly the same amount (3.8 and 3.6 cm) were determined. The initial retention factor estimates are plotted in *Figure A8.3* against vegetation densities obtained by linear interpolation. While there is some uncertainty in the assignment of values of Y, the general dependence of a_v on Y would probably be unchanged if the measured values of Y were to become known. The predicted values for the filtration model with $\mu = 1.5\ m^2\ kg^{-1}$ are shown as a solid line.

Two studies that determined average retention factors for ^{137}Cs and ^{90}Sr in fallout at widely separated locations were published in Health Physics in 1971 (van der Stricht et al. 1971). In the European study, monthly cuttings of herbage fed to cows were measured during a 6-month period in each year between 1961 and 1968. The total deposition during the growing season was measured and allowance was made for soil uptake. The best-fit values of the average retention fractions for ^{90}Sr and ^{137}Cs were 0.13 and 0.084, respectively (van der Stricht et al. 1971). The latter is quite similar to a value of 0.059 for ^{137}Cs found for a farm in Michigan in 1965 by fitting weekly measurements of deposition, rainfall, air concentration, and vegetation concentration to a deposition and retention model. A 14-day weathering half-life was assumed. The fitting process minimized the average fractional deviation between the predictions and measurements over a 6-month period (Pelletier and Voillequé 1971).

Mean values for retention of ^{137}Cs by alfalfa hay were reported by Ward et al. (1966). The estimates for the first cutting ranged from 0.21 to 0.37 and the mean of the long-term average values (0.27±0.07) was less than half the mean of values of a_v measured for individual storms during the early part of the growing season.

Estimates of retention of ^{90}Sr and ^{137}Cs by growing Kentucky bluegrass were reported for spring and summer of

Figure A8.3. Initial retention of ^{89}Sr in wet deposition as a function of vegetation density. Data from Menzel et al. (1963).



1965 (Krey and Fried 1966). The time between samples ranged from 2 to 6 weeks. Neither weathering nor dry deposition was considered, the latter in spite of the fact that covered plots were found to have more than half as much activity as those shielded from rain for three of seven measurement periods. Mean values of the retention factors for uncovered vegetation during the growing season were 0.13±0.10 and 0.072±0.086 for ^{90}Sr and ^{137}Cs, respectively. These values are quite similar to the other long-term values from van der Stricht et al. (1971) and Pelletier and Voillequé (1971).

Studies like these, which average over relatively long time periods, have resulted in lower estimates of the retention of wet deposition by vegetation. The initial retention factor, a_v, is not measured in these studies. A time-averaged retention is computed based on data on wet and dry deposition rates (or only the total) and vegetation concentration. The computed value inevitably reflects a variety of processes (including weathering) and their variations in time. The "retention factor" calculated in this way is not considered to be representative of a_v for ^{131}I in fallout particles.

Figure A8.4 shows the normalized retention factors for ^{90}Sr in fallout on four types of vegetables. The retention data for alfalfa during individual rainstorms at Fort Collins are also shown in the figure. For the alfalfa, it was necessary to estimate yields to obtain a_v; as for the vegetables (*Figure A8.3*), a linear growth rate was assumed. The solid line in the figure is the predicted normalized retention factor from *equation A8.6*. The comparison of the curve with the estimates of a_v is reasonable and provides encouragement that *equation A8.6* may represent a viable approach for estimating retention of fallout ^{131}I. However, *Figures A8.1, A8.2,* and *A8.4* all indicate that there is substantial variability among the measured values and that large uncertainties must be attached to estimates of a_v that are based on existing data.

A8.4.3. Retention by Vegetation of Sprays Containing Radionuclides

The principal alternative to measurements of the retention of wet deposition during rainstorms has been the use of man-made sprays of solutions of radiotracers or suspensions of labeled particles. Some of these studies were discussed in **Section A8.2** in connection with the development of the filtration model by Chamberlain. Two questions arise when the results are considered in connection with wet deposition of fallout, and fallout ^{131}I from the NTS in particular:

- Was the chemical form of the radionuclide similar to that expected in fallout from the NTS?
- Were the drop size distribution, fall velocity, and rainfall rate representative of those found in rainstorms?

When the answers to both questions are negative, the usefulness, for the present problem, of the estimates obtained is highly questionable. In general, the amount of "rainfall" applied in the form of a radioactive spray was quite low, less than 0.2 cm total. Drop size distribution and fall velocity were generally unspecified, but neither was likely to be representative of natural rain.

One of the several tests to measure ^{131}I transport in the milk-food chain performed by the Environmental Protection Agency at the NTS involved spraying a solution of ^{131}I as NaI (Douglas et al. 1971).

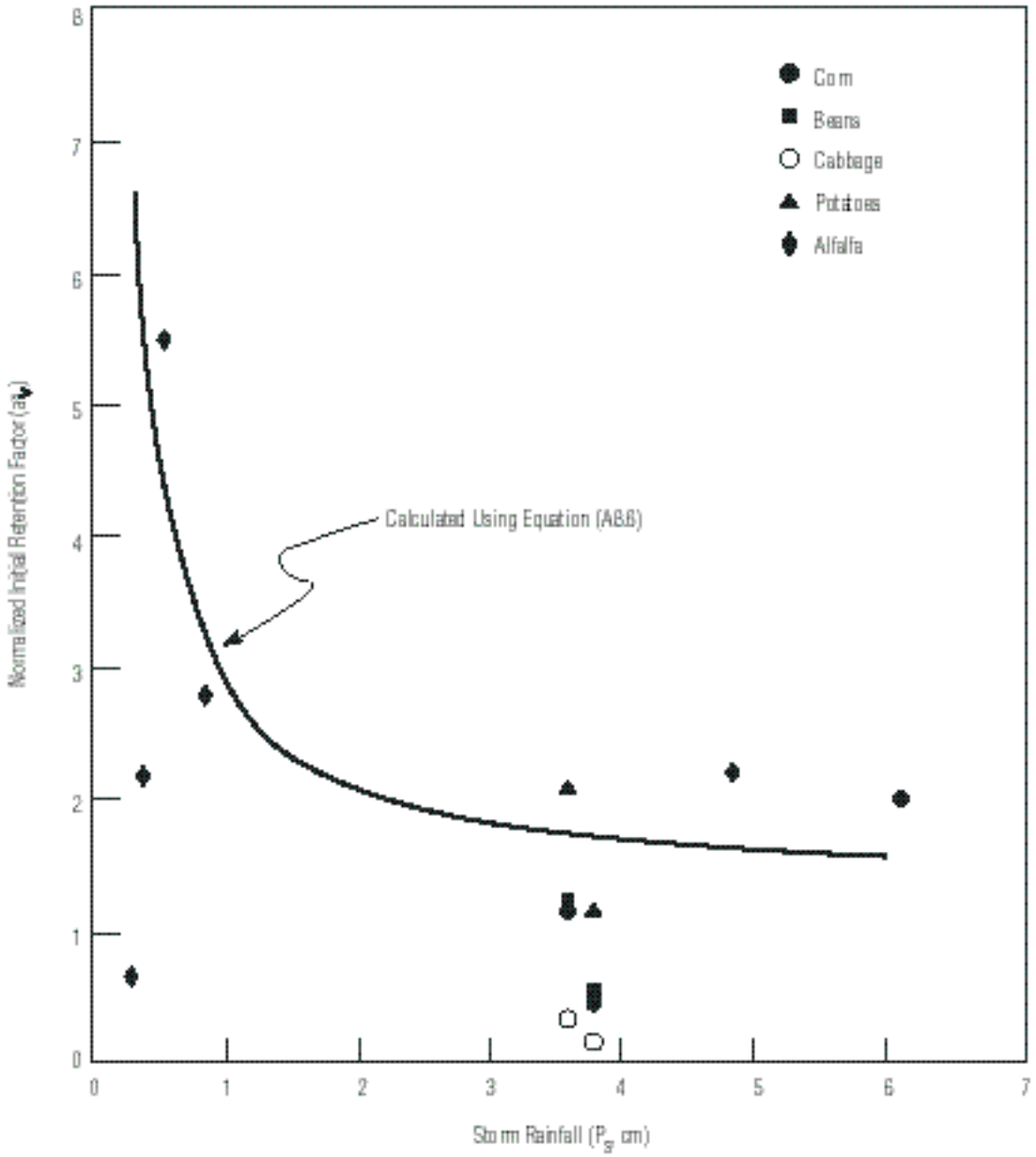
The ^{131}I would be present as iodide (I^-) in the solution. The total spray volume corresponded to a uniform rainfall of about 0.02 cm. The iodide was apparently tightly bound to the alfalfa; no washoff was detected, even for an (artificial) rainfall rate of more than l cm per hour. Neither the vegetation density nor an estimate of the initial retention was given in the report. However, Anspaugh (1987) has obtained an estimate of Y from one of the authors and has extrapolated the retention curve to obtain an estimate of 0.7 for the initial retention factor.

Anspaugh (1987) also cites a Swedish report describing a study by Edvarson and co-workers in which $Na^{131}I$ was sprayed onto a pasture. The equivalent rainfall was 0.008 cm. By assuming uniform ^{131}I metabolism among six cows, and that half the vegetation would be rendered unavailable due to trampling, an estimate of the initial retention of 0.2 was obtained.

Studies that involved spraying radiostrontium on several different pastures were conducted by Milbourn and his associates in the United Kingdom (Milbourn and Taylor 1965; Ellis et al. 1968). Although the pastures were diverse in history and use, similar retention results were obtained. The mean initial retention factor for seven measurements following imitation rainfalls of about 0.02 cm was 0.23±.05. Vegetation densities were low, ranging from 0.05 to 0.13 kg m^{-2}. The mean value of a^*_v is estimated to be 3.2 m^2 kg^{-1}. These data were used by Chamberlain in his development of the filtration model of initial retention. The effect of weathering was also measured and a best value of about 13 days was estimated, with a l-sigma range of 10 to 18 days.

As noted previously, Chamberlain found that fits to these data and some of his own results for labeled particles and solutions of radionuclides gave fairly consistent values for the parameter μ (m^2 kg^{-1}) in the filtration model. However, the data for fallout ^{90}Sr retention on vegetables are best approximated by a substantially smaller (factor of 2) value of μ (*Figure A8.3*).

Figure A8.4. Dependence of $a_{\downarrow}$ for fallout ^{90}Sr and ^{137}Cs on storm rainfall. Data from Menzel et al. (1963) and Ward et al. (1986).



Aarkrog (1969) studied the retention of a solution containing ^{85}Sr, ^{134}Cs, ^{54}Mn, and ^{144}Ce sprayed onto (artifically) high density plantings of grains (barley, oats, rye, and wheat). Similar results were found for all four radionuclides. The retention factors, measured 2 days after very light sprays of contaminated and then clean water, were nearly all above 0.4 and averaged 0.59 ± 0.14, 0.59 ± 0.13, 0.60 ± 0.12, and 0.55±0.15 for ^{54}Mn, ^{85}Sr, ^{134}Cs, and ^{141}Ce, respectively. The spray volume was small, giving $P_s \approx 0.008$ cm, and the clean water spray volume was the same. Vegetation densities ranged from 0.4 to 2.6 kg m^{-2}. Although no single species was tested at the full range of yields, the retention by rye, barley, and oats appears to peak between 1.0 and 1.5 kg m^{-2} and then declines, presumably because the plant mass is so thick that some stalks are protected from contamination. An awnless variety of wheat exhibited the lowest retention, but removal of awns from rye and other wheat varieties appeared to have little effect. Wheat plant densities were grouped at 0.3 to 0.4 and 0.8 to 1.0 kg m^{-2}, the overall mean retention was 0.52 ± 0.11, and no self-shielding could be discerned.

Retention by wheat was also studied by Middleton (1958, 1959), who found retention factors for carrier-free solutions of ^{89}Sr and ^{137}Cs which averaged 0.38 ± 0.11 and 0.39 ± 0.15, respectively. The spray volume for these results was even lower than that of Aarkrog, $P_s \approx 0.0001$ cm; no uncontaminated spray was used.

The complete retention density for rye grass was found to be about 0.6 kg m^{-2} by Kirchmann et al. (1966) using sprays of solutions containing ^{85}Sr and ^{134}Cs. Initial retentions of ^{85}Sr and ^{137}Cs were independent of tracer concentration and averaged 0.08 ± 0.01 and 0.05 ± 0.02, respectively for Y = 0.13 kg m^{-2}. The highest spray volume used decreased retention by about a factor of 2 for both ^{85}Sr and ^{137}Cs, but unfortunately, there is not enough information to determine the rainfall equivalents for the experiments.

Laboratory measurements to identify important variables affecting the initial retention by vegetation of ^{131}I in wet deposition have been performed at the Idaho National Engineering Laboratory (Maeck et al. 1984). Three series of tests were conducted using different chemical forms of ^{131}I added to a rain simulant. Preparation of the rain simulant was based on measurements of the contents of rain in remote, relatively unpolluted areas (Galloway et al. 1982). The three forms were CsI (I^- expected in the rain simulant), I_2 (I_2 and HOI expected in the rain simulant), and CH_3I (CH_3I expected in the simulant). The first of these forms is potentially the most relevant to wet deposition of fallout ^{131}I. The drops used had nominal diameters of 2.8 mm. The quantities applied were relatively low, equivalent to P_s <0.5 cm, and the drops did not fall at terminal velocity. That is, these measurements suffer from the same lack of realism as other experiments described in this section.

Initial retention by lettuce of ^{131}I applied as CsI varied from 0.16 to 0.56 of the total wet deposition in six experiments. Lower retention factors were measured for grass ($0.06 < a_v < 0.1$, 3 tests) and alfalfa ($a_v = 0.02$, 1 test). Vegetation densities ranged from 0.06 to 0.5 kg/m^2 in the 10 experiments. Although the values of a_v generally increased with Y, the correlation was not strong ($r^2 = 0.12$). It was found that results with an acidified rain simulant were comparable to the normal simulant. Retention of contaminated drops was lower when the contamination event was preceded by exposure to uncontaminated drops. This agrees qualitatively with the idea proposed by Horton (1919).

As indicated previously, the relevance of all the results in this section to the problem of predicting the initial retention of ^{131}I in wet deposition of fresh fallout, while uncertain, is believed to be small. In the absence of new information, reliance should be placed on the results of experiments described in **Sections A8.4.1** and **A8.4.2.**

A8.4.4. Retention by Vegetation of ^{7}Be

Some results of measurements of the behavior of ^{7}Be in wet deposition were reviewed. Olsen et al. (1985) measured the deposition of ^{7}Be and the resulting inventories of ^{7}Be in soil and vegetation at coastal sites and at Oak Ridge. Monthly total deposit on fluxes were measured. The ^{7}Be was found to all be in the liquid phase, operationally defined by passage through a filter with a pore size of 0.45 μm. Laboratory experiments, theoretical analyses, and the results of other workers all supported the belief that the isotope was present in rain water as $^{7}Be^{++}$. A comparison of the total ^{7}Be deposition and the ^{7}Be wet deposition at Norfolk, Virginia indicated that dry deposition contributed less than 10% of the total. Statistical evaluation of data on total ^{7}Be deposition and the associated monthly rainfalls led to the conclusion that dry deposition accounted for 30 ± 16% and 19 ± 16% of the total deposition at Norfolk and Oak Ridge, respectively. These estimates were believed to be biased on the high side because of the decrease in the concentration in rain as precipitation increases. Measurements of ^{90}Sr showing such a decrease were reported by Krey and Toonkel (1977) (**Section A8.3.1**). Calculation of the dry deposition rate using a mean deposition velocity of 0.23 cm s^{-1} to grasses (Bondietti et al. 1984) and the average ^{7}Be concentration in air at Oak Ridge, 0.09 pCi m^{-3} yields 0.054 pCi cm^{-2} $month^{-1}$, about 15% of the total.

The data on ^{7}Be in soil and vegetation at Oak Ridge and several marshes given by Olsen et al. (1985) were analyzed in terms of a simple model. Uptake of deposited ^{7}Be from the soil was expected to be small. The value of the dimensionless uptake ratio, B_v, for Be is estimated to be 4.2×10^{-4} (NRC 1977). Because of the low value of B_v, soil uptake was not considered. The equations used were:

$$\frac{dC_v}{dt} = aD - \lambda_e C_v \qquad (A8.8)$$

$$\frac{dC_s}{dt} = (1 - a)\ D + \lambda_w C_v - \lambda C_s \qquad (A8.9)$$

where:

- a is the average retention by grass
- D is the total (wet and dry) deposition rate (pCi cm^{-2} s^{-1})
- λ_e is the effective removal rate constant (s^{-1}), equal to $(\lambda + \lambda_w)$
- C_s is the concentration (pCi m^{-2}) of ^{7}Be in soil

The symbols C_v, λ, and λ_w were defined following *equation A8.1.*

The results of measurements of ^{7}Be at the Oak Ridge and three marsh sampling locations are shown in *Table A8.1.* When the grass and soil inventories are compared, it seems clear that either the retention (a) is high or removal of ^{7}Be by weathering is a slow process. Bounding estimates for the average initial retention and weathering half-life can be made from these data if it is assumed that the samples were representative of an equilibrium situation.

When equilibrium is reached, the concentrations in vegetation and soil would be:

$$C_{ve} = \frac{aD}{\lambda_e} \qquad (A8.10)$$

$$C_{se} = \frac{D}{\lambda} - \frac{aD}{\lambda_e} \qquad (A8.11)$$

respectively, and the ratio of the two activities would be:

$$\frac{C_{se}}{C_{ve}} = \frac{\lambda_e - a\lambda}{a\lambda} \qquad (A8.12)$$

Table A8.1. Estimates of minimum average initial retention and weathering half-life for ^{7}Be from inventory measurements.

Location and Date	Measured ^{7}Be Inventories (pCi cm^{-2})		Estimates based on *equation A8.12*	
	Grass	Soil	(If $\lambda_w = 0$) a	(If a = 1) T_w (days)
Oak Ridge, ORNL Soil W-3 (7-3-84)	1.48 ± .030	34 ± .05 (0.64)[a]	0.81 (0.70)	230 (120)
James River Marsh (8-12-82)	1.15 ± .12	0.65 ± .11	0.64	120
Delaware Marsh (7-2-82)	0.33 ± .04	0.23 ± .06	0.60	76
Wallops Island Marsh (1-3-85)	0.77 ± .07	1.05 ± .11 (0.39)[a]	0.42 (0.66)	39 (110)

[a] Computed by taking the difference between the vegetation concentration and the estimated total inventory based on deposition measurements. At ORNL, the estimated inventory was 2.12 pCi cm^{-2}, and at Wallops Island it was 1.16 pCi cm^{-2}.

The last columns of *Table A8.1* show the minimum values of a (assuming no removal by weathering) and of the removal half-life (assuming the maximum initial retention, a = 1). The estimates suggest that the behavior of ^{7}Be differs greatly from the behavior of fallout particles. Long-term average values of retention of fallout ^{137}Cs and ^{90}Sr were typically about 0.1 (**Section A8.4.2**). These results suggest that there must be another source of ^{7}Be in vegetation. Uptake from the soil or rainsplash of soil particles onto vegetation with avid retention seem to be the only alternatives. As noted above, soil uptake was expected to be small.

Measurements by Mahoney (1984) of the initial retention by clover of ^{7}Be during a rainstorm at Oak Ridge in early May 1983 yielded a value of 0.18, substantially lower than the minimum values derived in *Table A8.1.* The clover density was 0.077 kg m^{-2}; therefore, a^*_v was 2.3 m^2 kg^{-1} for P_s = 1.3 cm. Three later measurements for fescue and clover integrated over three storms (the rainfall for the principal ^{7}Be deposition was P_s = 3.6 cm) were reported to yield initial retention estimates of 0.16 to 0.18. However, data tabulated in the report suggest possible mathematical errors and that the values of initial retention factor may have been 0.02 for fescue and 0.02 and 0.06 for clover. Until the inconsistencies can be clarified, it is not possible to say which values are the correct ones.

Two experiments (Mahoney 1984) to measure the weathering half-life of ^{7}Be that was sprayed onto fescue plots at Oak Ridge National Laboratory yielded average values of 36.5 days (during an 80-day period in winter) and 38.5 days (during a 70-day period in spring). During the winter measurements, most of the decrease in concentration was observed during the first 2 weeks. The weathering half-life for that period, estimated from data in the report, was about 6 days.

Mahoney (1984) did find rapid adsorption of soluble ^{7}Be by plant leaves. Freshly harvested fescue and bean leaves were exposed to solutions containing Be^{++}, $^{137}Cs^{+}$, and $^{131}I^{-}$. The leaves were then rinsed prior to analysis. The two positive ions were adsorbed in a similar manner with observed $^{137}Cs/^{7}Be$ ratios in exposed vegetation ranging from 0.8 to 2.4. The ratios of $^{7}Be^{++}$ to $^{131}I^{-}$ ranged from l0 to 40 in one experiment and from 50 to 150 in another. The $^{137}Cs^{+}$ to $^{131}I^{-}$ ratios ranged from 50 to 250 (Mahoney 1984). These results are qualitatively similar to those of Angeletti and Levi (1975) who found substantially greater retention by vegetation of Sr^{++} than of I^{-} when solutions were sprayed on the plants. Mahoney (1984) found that about 1/3 of the total adsorption of both ^{7}Be and ^{131}I occurred within 3 minutes in one experiment ($^{7}Be^{++}/^{131}I^{-}$ ranged from 10 to 40). In the other experiments, the first measurements were not made until 30 minutes after the start of exposure). These results suggest that $^{7}Be^{++}$ in rainwater would be promptly bound to plant leaves and could lead to high initial retentions. They also suggest that $^{7}Be^{++}$ is not a good analog for studying the retention by vegetation of ^{131}I present in rainwater as I^{-}.

One measurement of uptake of ^{7}Be injected into soil in which fescue was growing was reported by Mahoney (1984). It was found that only 0.13 ± 0.04% of the injected activity was present in the vegetation after a 2-month growth period. Although a direct comparison with the reported value of B_v (NRC 1977) is not possible, this result indicates that soil uptake is not adequate to account for the discrepancies implied by analysis of the field data (*Table A8.1*).

When the average values of the retention fraction and weathering half-life measured for fescue at Oak Ridge are substituted into *equation A8.12*, the predicted ratio of the ^{7}Be inventory in soil to that in vegetation is found to be about 13. This ratio is about ten times larger than the largest of the ratios computed from the measured inventories shown in *Table A8.1.* If the estimated weathering half-life is correct, equilibrium will be achieved fairly rapidly. At Oak Ridge (Olsen et al. 1985), the estimated total inventories for the 4 months of April to July 1984 were 1.85, 2.38, 2.12, and 2.17 pCi m^{-2}, respectively. Thus, the assumption of equilibrium in deriving *equation A8.12* does not appear to be invalid for that location. Estimates of the ^{7}Be inventory for the months preceding the measurements at the coastal locations were not given.

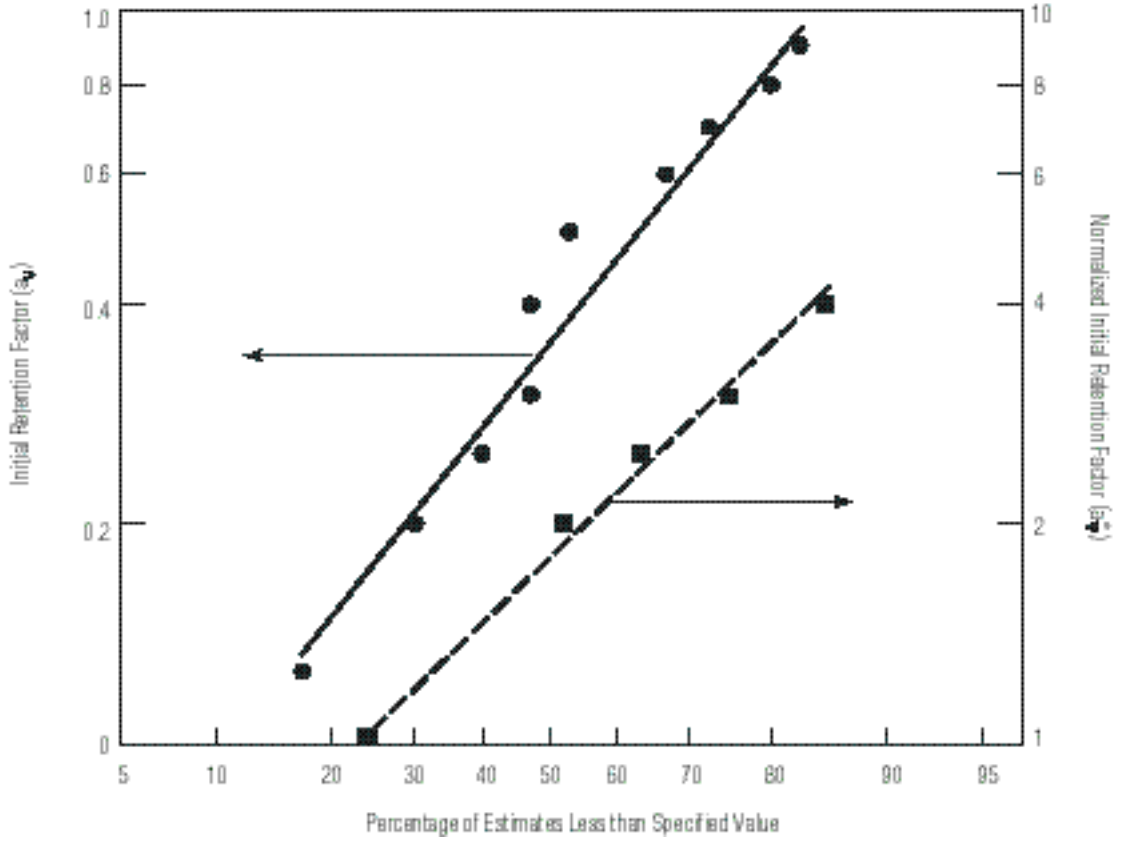
In a study of deposition of Chinese fallout particles onto tree canopies, Russell and Choquette (1976) found wet deposition to be the predominant transport mechanism from atmosphere to leaves. A long retention period was observed with estimated half-lives of 50 to 200 days, depending upon the leaf type. The transport of ^{7}Be was also measured. Relative to ^{141}Ce, only 20% of the ^{7}Be in rainfall was fixed on tree leaves. The reason for this observation is not known but may be related to differences in particle size, solubility, or binding to the leaf surface.

A8.5. SUMMARY OF RELEVANT DATA

The information considered most relevant to the task of assessing the initial retention by vegetation of ^{131}I in wet deposition of fallout from the NTS is that discussed in **Sections A8.4.1** and **A8.4.2.** The available data and conceptual evaluations both suggest that the initial retention factor is dependent upon both the density of the vegetation (Y, kg m^{-2}) and upon the amount of rainfall. Other related parameters (total leaf area, leaf surface characteristics, rainfall rate, rainfall sequence, and so on) clearly enter into the observed interception and initial retention processes, but examination of the processes at that level of detail is beyond the scope and needs of the current dose evaluation effort.

Estimates of the initial retention factor, a_v, based upon field measurements of wet deposition onto pasture grass range from < 0.09 (Chinese fallout in the midwestern U.S.) to 1.0 (Russian fallout in the United Kingdom). Thirty estimates are available from Peirson and Keane (1962), Weiss et al. (1974), and Voillequé et al. (1981). The mean of these values (with sample standard deviation)is 0.45 ± 0.32. Chamberlain and Chadwick (1966) found an average of 0.5 ± 0.10 for a

Figure A8.5. Distributions of a_v and a^*_v for wet deposition of ^{131}I in fallout.



comparable number of measurements. Five measurements for fallout ^{137}Cs in wet deposition onto alfalfa ranged from 0.26 to 0.83 with a mean of 0.60 ± 0.21 (Ward et al. 1965).

A normalized initial retention factor can be used to incorporate the approximately linear dependence upon vegetation density of the filtration model developed by Chamberlain (1970). Twenty values of $a^*_v = a_v/Y$ obtained from measurements of Chinese fallout ^{131}I in the midwestern United States range from 0.56 to 5.5 $m^2\ kg^{-1}$ with a mean of 2.0 ± 1.6 $m^2\ kg^{-1}$. Inclusion of the five results for fallout ^{137}Cs deposited on alfalfa yields a mean of 2.1 + 1.6 $m^2\ kg^{-1}$.

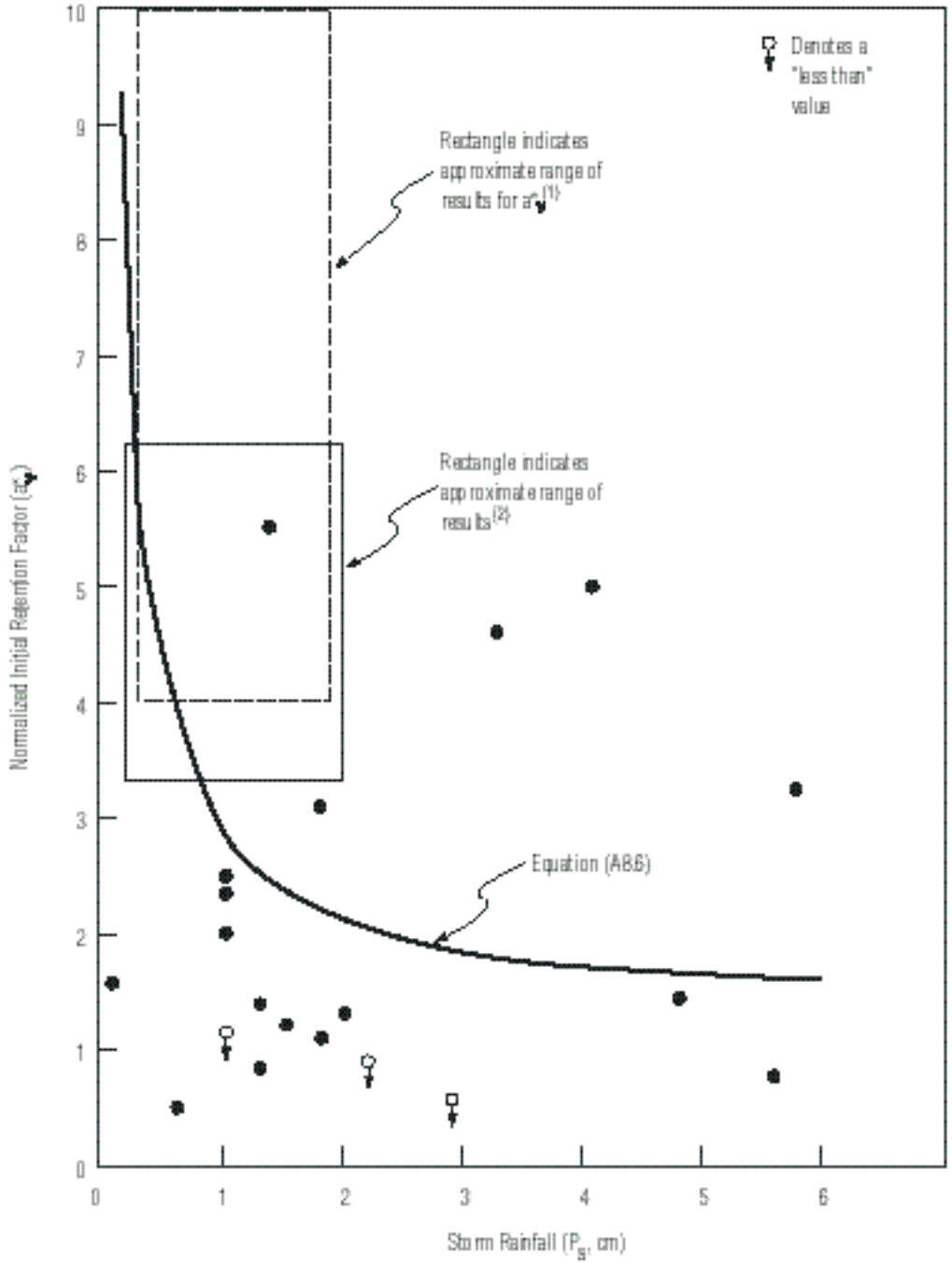
Figure A8.5 shows the distributions of the 30 values of a_v and 20 values of a^*_v. Use of the normalized initial retention factor does reduce the variability somewhat. The median value of a_v is estimated to be 0.35 with a geometric standard deviation of 2.9. The estimated median a^*_v is 1.8 $m^2\ kg^{-1}$, with a geometric standard deviation of 2.3.

The available data on the initial retention of fallout ^{131}I and of particulate fallout nuclides in wet deposition suggest that a model similar to that proposed by Horton (1919) may be used to estimate the effect of differences in rainfall amount. *Figure A8.6* shows the suggested relationship (*equation A8.6*) together with values of a^*_v from the midwestern United States and ranges of a^*_v for the United Kingdom. These ranges are based upon the estimated ranges of storm rainfall (0.2 to 2 cm) and of autumn vegetation densities (0.08 to 0.15 kg m^{-2}). While the comparison between *equation A8.6* and the estimates of a^*_v are not particularly satisfying, other approximations that might be used are likely to have comparable deficiencies when attempting to account for the variety of measurement results.

A8.6. CONCLUSIONS

The initial retention by pasture vegetation of ^{131}I in wet deposition at locations in the United States is an important factor in the assessment of the thyroid doses received from NTS fallout. The projected doses from wet deposition are proportional to the initial retention factor, a_v. Wet deposition will be the most significant transport process for many parts of the country and perhaps for the collective thyroid dose.

Figure A8.6. Estimated values of $a\hat{v}$ as a function of storm rainfall. Data from (1) Peirson and Keane (1962) and (2) Chamberlain and Chadwick (1966).



Data collected during 1961 and 1962 and more recently suggest that ^{131}I in fresh fallout is primarily associated with particulate debris. During the few days when radioactivity would have been transported from the NTS to other locations in the United States, a minimum of 60%, and as much as 90%, of the ^{131}I activity was in particulate form. Inorganic forms would dominate the gaseous fraction with at most one-third of that component present as organic iodides soon after detonation. At least half of the ^{131}I reaching the ground in rainwater would be contained in scavenged particles having diameters between 1 and 20 μm. Part of the remainder could be composed of submicron particles.

Field measurements of wet deposition of ^{131}I and other radionuclides in fallout and tests involving various types of fallout simulants indicate the initial retention factor depends upon both the vegetation density (Y, kg m^{-2}) and the total amount of rainfall during a storm (P_s, cm). Use of the normalized initial retention factor ($a^*_v = a_v/Y$) reduces the variability of the field measurement results. The median value of 30 estimates of a^*_v was 0.35, with a geometric standard deviation of 2.9. The median of 20 estimates of a^*_v was 1.8 m^2 kg^{-1}, with a geometric standard deviation of 2.3.

Detailed evaluation of variations due to changes in precipitation rate during a storm is beyond the needs and the resources of the fallout dose reassessment effort. However, it is desirable to know the dependence of the initial retention factor on the total storm rainfall. The approach suggested by Horton was used. Existing data for fallout were used to develop a predictive equation for the normalized initial retention factor: $a^*_v = (S/P_S) + E$, where S and E are constants related to rainfall storage capacity and evaporation during a storm (**Section A8.4.1**). In the absence of other information, this equation appears to provide a reasonable estimate of the dependence upon rainfall. The alternative (again in the absence of new measurement results) would be to use the median value of a^*_v.

Most wet deposition simulation experiments have been conducted under extremely light spray conditions. These tests are not considered reliable indicators of fallout ^{131}I behavior for that reason and because the tracer forms were not reflective of fresh fallout containing ^{131}I.

The use of ^{7}Be as an analog for ^{131}I in fresh fallout is not considered to be a reliable alternative. At most, half of the ^{131}I would be expected to be in solution, compared with all of the ^{7}Be. Further, the adsorption by leaves of $^{7}Be^{++}$ from solution appears to be much greater than the adsorption of $^{131}I^-$ by the same leaves. The observed behavior of ^{7}Be in wet deposition onto grass and soil differs greatly from that deduced for radionuclides in fallout particles. The fact that both ^{7}Be and ^{131}I are both poorly retained by gummed film as the rainfall amount increases is considered to reflect water saturation of the film surface and runoff rather than an inherent similarity in retention of the two nuclides.

Field experiments performed to determine the dependence of a_v on vegetation density and rainfall parameters should employ ^{131}I as iodide and iodate in solution and particles with diameters of up to about 20 μm with tightly bound radionuclide labels. The spray system used should generate a realistic simulant of natural rain. The raindrop size spectrum as a function of rainfall intensity needs to be well characterized.

REFERENCES

Aarkrog, A. On the direct contamination of rye, barley, wheat, and oats with ^{85}Sr, ^{134}Cs, ^{54}Mn, and ^{141}Ce. Rad. Bot. 9: 357; 1969.

Angeletti, L.; Levi, E. Retention and adsorption of foliar applied iodine and strontium on rye grass and clover. Paper presented at the European Congress of the International Radiation Protection Association, Amsterdam; 1975.

Anspaugh, L. R. Retention by vegetation of radionuclides deposited in rainfall—A literature summary. UCRL-53810 Lawrence Livermore National Laboratory, Livermore, CA 1987.

Beck, H. L. Estimates of fallout from Nevada weapons testing in the western United States based on gummed-film monitoring data. EML-433. New York, NY: Environmental Measurements Laboratory, U.S. Department of Energy; 1984.

Beck H. L. Personal communication; 1986.

Benson, P.; Cleit, C.E.; Leventhal, L. Physical characteristics of single particles from high-yield air bursts. In: Radioactive fallout from nuclear weapons tests (edited by A.W. Klement, Jr.), AEC Symposium Series 5, CONF-765. Springfield, VA: National Technical Information Service; 1965.

Benson, P.; Cleit, C.E.; Leventhal, L. Radiochemical fractionation characteristics of single particles from high-yield air bursts. In: Radioactive fallout from nuclear weapons tests (edited by A.W. Klement, Jr.), AEC Symposium Series 5, CONF-765. Springfield, VA: National Technical Information Service; 1965.

Bergström, S.O.W. Transport of fallout ^{131}I into milk. In: Radioecological concentration processes (edited by B. Aberg and F.P. Hungate). Oxford: Pergamon Press; 1967.

Bergström, S.O.W.; Gyllander, C. Iodine-131 in grass and milk correlated with releases from the Studsvik Research Station. In: Environmental contamination by radioactive material. Vienna: International Atomic Energy Agency; 1969.

Black, S. C.; Potter, G. D. Historical perspectives on selected health and safety aspects of nuclear weapons testing. Health Phys. 51:17, 1986.

Burgy, R. H.; Pomeroy, C. R. Interception losses in grassy vegetations. Trans. AGU, 80:1095, 1958.

Bondietti, E. A.; Hoffman, F. O.; Larsen, I. L. Air-to vegetation transfer rates of natural submicron aerosols. J. Environ. Radioactivity 1: 5, 1984.

Chadwick, R. C.; Chamberlain, A.C. Field loss of radionuclides from grass. Atmos. Environ., 4:51, 1969.

Chamberlain, A.C.; Dunster, H.J. Deposition of radioactivity in northwest England from the accident of Windscale. Nature 182: 629, 1958.

Chamberlain, A.C.; Chadwick, R.C. Transport of iodine from atmosphere to ground. Tellus 18:226, 1966.

Chamberlain, A.C. Transport of Lycopodium spores and other small particles to rough surfaces. Proc. Roy. Soc. A296: 1967.

Chamberlain, A.C. Interception and retention of radioactive aerosols by vegetation. Atmos. Environ. 4:57, 1970.

Crocker, G.R.; Kawahara, F.K.; and Freiling, E.C. Radiochemical data correlations on debris from silicate bursts. In: Radioactive fallout from nuclear weapons tests (edited by A.W. Klement, Jr.), AEC Symposium Series 5, CONF-765. Springfield, VA: National Technical Information Service; 1965.

Douglas, R.L.; Black, S.C.; Barth, D. S. ^{131}I transport through the air-forage-cow-milk system using an aerosol mist. SWRHL-43R. Las Vegas, NV: Southwestern Radiological Health Laboratory, U. S. Environmental Protection Agency; 1971.

Drevinsky, P.J.; Pecci, J. Size and vertical distributions of stratospheric radioactive aerosols. In: Radioactive fallout from nuclear weapons tests (edited by A.W. Klement, Jr.), AEC Symposium Series 5, CONF-765. Springfield, VA: National Technical Information Service; 1965.

Eggleton, A.E.J.; Atkins, D.H.; Cousins, L.B. Chemical and physical nature of fallout ^{131}I and carrier-free ^{131}I released in air—An Abstract. Health Phys. 9:1111, 1963.

Ellis, F.B.; Mercer, E.R.; Milbourn, G.M. The contamination of grassland with radioactive strontium-II. Effect of time and cultivation on the levels of strontium-90 in herbage. Rad. Bot. 8:269, 1968.

Engelmann, R.J.; Slinn, W.G.N. (Coordinators). Precipitation scavenging (1970). AEC Symposium Series 22, CONF-700601 Springfield, VA: National Technical Information Service; 1970.

Engelmann, R.J. Scavenging prediction using ratios of concentrations in air and precipitation. In: Precipitation scavenging (l970).(R.J. Engelmann and W.G.N. Slinn), AEC Symposium Series 22, CONF-700601. Springfield, VA: National Technical Information Service; 1970.

Engelmann, R.J.; Sehmel, G.A. (Coordinators). Atmosphere-surface exchange of particulate and gaseous pollutants. ERDA Symposium Series 38, CONF-740921. Springfield, VA: National Technical Information Service; 1976.

Friedlander, S.K.; Pasceri, R.E. Measurements of the particle size distribution of the tropospheric aerosol with the rotating-disk Sampler. In: Radioactive fallout from nuclear weapons tests (edited by A.W. Klement, Jr.), AEC Symposium Series 5, CONF-765. Springfield, VA: National Technical Information Service; 1965.

Freiling, E. C.; Crocker,G.R.; Adams,C.E. Nuclear-debris formation. In: Radioactive fallout from nuclear weapons tests (edited by A.W. Klement, Jr.), AEC Symposium Series 5, CONF-765. Springfield, VA: National Technical Information Service; 1965.

Gatz, D.F. Scavenging ratio measurements in METROMEX. In: Precipitation Scavenging (l974). (coordinated by R.W. Beadle and R.G. Semonin), ERDA Symposium Series 41, CONF-741003. Springfield, VA: National Technical Information Service, 1977.

Galloway, J.N.; Keene, W.C.; Likens, G.E.; Miller, J.M. The composition of precipitation in remote areas of the world. J. Geophys. Res. 87: 8771, 1982.

Georgii, H.W.; Pankreth, J. Deposition of atmospheric pollutants. Boston: D. Reidel Publishing Company; 1982.

Glasstone, S. The effects of nuclear weapons, revised edition. Washington, DC: Superintendent of Documents; 1964.

Hoffman, L.G.; Keller J. H.; and Voillequé P. G. 1982, Wet Deposition Processes for Radioiodines, (Washington, DC: U. S. Nuclear Regulatory Commission), NUREG7CR-2438.

Hoffman, F. 0. A reassessment of the deposition velocity in the prediction of the environmental transport of radioiodine from air to milk. Health Phys. 32: 437; 1977.

Horton, R.E. Rainfall Interception. Mon. Weath. Rev. 49:603, 1919.

Huff, F.A. Radioactive rainout relations on densely gauged sampling networks. In: Radioactive fallout from nuclear weapons tests (edited by A.W. Klement, Jr.), AEC Symposium Series 5, CONF-765. Springfield, VA: National Technical Information Service; 1965.

Hull, A.P. Vegetation retention and vegetation-milk ratios of fallout ^{131}I. Health Phys. 9: 1173; 1963.

Keisch, B.; Koch, R.C. Physical and chemical states of iodine in fallout. Status report No. 2, AEC Report NSEC-84. Pittsburgh, PA: Nuclear Science and Engineering Corporation; 1963.

Keller, J.H.; Hoffman, L.G.; Voillequé, P.G. Laboratory measurements of parameters affecting wet deposition processes for radioiodines. NRC Report NUREG/CR-2438. Washington, DC: U.S. Nuclear Regulatory Commission; 1982.

Kins L. Temporal variation of chemical composition of rainwater during individual precipitation events. In: Deposition of atmospheric pollutants. (Edited by H.-W. Georgii and J. Pankreth) Boston: D. Reidel Publishing Company; 1982.

Kirchmann, R.; Fagnard, E.; van Puymbroeck, S. Studies on foliar contamination by radiocesium and radiostrontium. In: Radioecological concentration processes. (B. Aberg and F. P. Hungate, editors). New York: Pergamon Press; 1966.

Kirchner, T.B.; Whicker F. W. Validation of PATHWAY, a simulation model of the transport of radionuclides through agroecosystems. Ecological Modelling 22: 21; 1983.

Koch R.C.; Keisch, B. Physical and chemical states of iodine in fallout. Status report No. 1, AEC Report NSEC-79. Pittsburgh, PA: Nuclear Science and Engineering Corporation; 1962.

Krey, P.W.; Toonkel, L.E. Scavenging ratios. In: Precipitation scavenging (1974), (Coordinated by R.G. Semonin and R.W. Beadle). ERDA Symposium Series 41, CONF-741003. Springfield, VA: National Technical Information Service; 1977.

Krey, P.W.; Fried, R.E. Long range fallout from Sedan and Small Boy shots. In: Radioactive fallout from nuclear weapons tests (edited by A.W. Klement, Jr.), AEC Symposium Series 5, CONF-765. Springfield, VA: National Technical Information Service; 1965.

Lassey, K.R. The interception and retention of aerosols by vegetation. I. The formulation of a filtration model. Atmos. Environ. 16: 13; 1982.

Lassey, K.R. The interception and retention of aerosols by vegetation. II. A filtration model description for various aerosols on grassland. Atmos. Environ. 17: 2303; 1983.

Lassey, K.R. Grassland dynamics of radioiodine: a filtration model simulation. J. Environ. Radioact. 1: 29;1984.

Lockhart, L.B., Jr.; Patterson, R.L., Jr.; Saunders, A.W., Jr. Distributions of airborne radioactivity with particle size. In: Radioactive fallout from nuclear weapons tests (edited by A.W. Klement, Jr.), AEC Symposium Series 5, CONF-765. Springfield, VA: National Technical Information Service; 1965.

Loysen, P. Particle size distribution of stratospheric aerosols at 110,000 ft. In: Radioactive fallout from nuclear weapons tests (edited by A.W. Klement, Jr.), AEC Symposium Series 5, CONF-765. Springfield, VA: National Technical Information Service; 1965.

MacKenzie, E.H. A. J.; Menzel, R.G.; Roberts, H.,dr.; and Stewart. Strontium-90 Accumulation on Plant Foliage During Rainfall. Science, 142, 576; 1963.

Maeck, W.J.; Honkus, R.J.; Keller, J.H; Voillequé, P.G. Laboratory measurements of parameters affecting wet deposition of methyl iodide. NRC Report NUREG/CR-40l4. Washington, D.C.: U.S. Nuclear Regulatory Commission; 1984.

Mahoney, L.A. Beryllium-7 deposition to terrestrial vegetation in Tennessee. Draft thesis. Bowling Green, KY: Western Kentucky University; 1984.

Menzel, R.G.; Roberts, H., Jr.; Stewart, E.H.; MacKenzie, A.J. Strontium-90 accumulation on plant foliage during rainfall. Science 142: 576;1963.

Middleton, L.J. Absorption and translocation of strontium and cesium by plants from foliar sprays. Nature 181:1300; 1958.

Middleton, L.J. Radioactive strontium and cesium the edible parts of crop plants after foliar contamination. Inter. J. Radiat. Biol. 4: 387; 1959.

Milbourn, G.M.; Taylor, R. The contamination of grassland with radioactive strontium - I. The initial retention and loss. Rad. Bot. 5: 337; 1965.

Miller, C.W. Validation of a model to predict aerosol interception by vegetation. In: Biological implications of radionuclides released from nuclear industries. Vienna: International Atomic Energy Agency; 1979.

Miller, C.W. An analysis of measured values for the fraction of a radioactive aerosol intercepted by vegetation. Health Phys. 38: 795; 1980.

NRC (Nuclear Regulatory Commission). Calculation of annual doses to man from routine releases of reactor effluents for the purpose of evaluation compliance with 10 CFR 50, Appendix I. Regulatory Guide 1.109, rev. 1. Washington, DC: U. S. Nuclear Regualtory Commission; 1977.

Olsen, C.R.; Larsen, I.L.; Lowry, P.D.; Cutshall, N.H.,; Todd, D.F; and Wong, G.T.F; Casey, W.H. Atmospheric fluxes and marsh-soil inventories of ^{7}Be and ^{210}Pb. J. Geophys. Res. 90: 10487, 1985.

Peirson, D.H.; Keane, J.R. Characteristics of early fall-out from the Russian nuclear explosions of 1961. Nature 196: 801; 1962.

Pelletier, C.A.; Voillequé, P.G. The behavior of ^{137}Cs and other fallout radionuclides on a Michigan dairy farm. Health Phys. 21:777; 1971.

Perkins, R.W. Physical and chemical form of ^{131}I in fallout. Health Phys. 9:1113; 1963.

Perkins, R.W.; Thomas, C.W.; Nielsen, J.M. Measurements of airborne radionuclides and determination of their physical characteristics. In: Radioactive fallout from nuclear weapons tests (edited by A. W. Klement, Jr.). AEC Symposium Series 5, CONF-765. Springfield, VA: National Technical Information Service; 1965.

Peters, L.N.; Witherspoon, J.P. Retention of 44-88 μm simulated fallout particles by grasses. Health Phys. 22: 261; 1972.

Pruppacher, H.R.; Semonin, R.G.; Slinn, W.G.N. Precipitation scavenging, dry deposition, and resuspension. Two volumes. New York: Elsevier; 1983.

Reiter, E.R. Atmospheric transport processes, part 4: radioactive tracers. DOE Critical Review Series, TID-27224. Springfield, VA: National Technical Information Service; 1978.

Riedel, H.; von Gadow, A.; Gesewsky, P. Investigation into the occurrence of radioiodine in air, precipitation, grass, cow and goat milk following the Chinese atomic bomb test of September 26, 1976. STH-Report 3/77. Munich, FRG: Institute for Radiation Hygiene of the Federal Health Office, 1977.

Russell, I.J.; Choquette, C.E. Scale factors for foliar contamination by stratospheric sources of fission products in the New England area. In: Atmosphere-Surface Exchange of Particulate and Gaseous Pollutants, ERDA Symposium Series 38, CONF-740921. Springfield, VA: National Technical Information Service, 1977.

Semonin, R.G.; Beadle, R.W. (Coordinators). Precipitation Scavenging (1974). ERDA Symposium Series 41, CONF-741003. Springfield, VA: National Technical Information Service; 1977.

Slinn, W.G.N. Precipitation scavenging: some problems, approximate solutions, and suggestions for future research. In: Precipitation scavenging (1974) (coordinated by R. G. Semonin and R. W. Beadle). ERDA Symposium Series 41, CONF-741003. Springfield, VA: National Technical Information Service; 1977.

van der Stricht, E.; de Bortoli M., Gaghone P. Predictions of 90Sr levels in milk on the basis of deposition values. Health Phys . 21: 217, 1971.

Voillequé, P.G. Iodine species in reactor effluents and in the environment. EPRI report NP-1269. Palo Alto, CA: Electric Power Research Institute; 1979.

Voillequé, P.G.; Kahn, B.; Krieger, H.L.; Montgomery, D.M.; Keller, J.H.; Weiss, B.H. Evaluation of the air-vegetation-milk pathway for ^{131}I at the Quad Cities Nuclear Power Station. NRC Report NUREG/CR-1600. Washington, D.C.: U.S. Nuclear Regulatory Commission; 1981.

Ward, G.M.; Johnson, J.E.; Wilson, D.W. Deposition of fallout cesium-137 on forage and transfer to milk. Public Health Reports, 81:639; 1966.

Ward, G.M.; Johnson, J.E.; Stewart, H.F. Cesium-137 passage from precipitation to milk. In: Radioactive fallout from nuclear weapons tests (edited by A.W. Klement, Jr.), AEC Symposium Series 5, CONF-765. Springfield, VA: National Technical Information Service; 1965.

Weiss, M.M. A study of the airborne radioactivity at Brookhaven National Laboratory from the Nevada tests. AEC report BNL 352 (T-39). Upton, NY: Brookhaven National Laboratory; 1953.

Weiss, B.H.; Voillequé, P.G.; Keller, J.H.; Kahn, B.; Kreiger, H.L.; Martin, A.; Phillips C. R. Detailed Measurement Of ^{131}I in air, vegetation, and milk around three operating reactor aites. In: Environmental surveillance around nuclear installations, Volume I. Vienna: International Atomic Energy Agency; 1974.

Wilson, D.W.; Ward, G.M.; Johnson, J.G. Fallout ^{137}Cs: direct aerial transfer as an important source of foliar deposition. Rad. Bot. 7: 312; 1967.

Wilson, D.A. A mathematical model of the transport of cesium-137 from fallout to milk. Ph.D. Dissertation. Fort Collins, CO: Colorado State University; 1968.

Witherspoon, J.P.; Taylor, F.P., Jr. Interception and retention of a simulated fallout by agricultural plants. Health Phys. 19:483; 1970.

Witherspoon, J.P.; Taylor, F.P., Jr. Interception and retention of 1-44 μ simulated fallout particles by soybean and sorghum plants. Health Phys. 21:673; 1971.

Zimbrick, J.D.; Voillequé, P.G. Controlled environmental radioiodine tests at the National Reactor Testing Station. AEC report IDO-12065. Idaho Falls, ID: U.S. Atomic Energy Commission; 1969.